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Data Reduction Techniques for Aerosol Size Distribution Measuring Instruments

Minnesota Univ.
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Abde Kapadia.

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Data reduction methods for the electrical aerosol analyzer (EAA) and the diffusion battery+condensation nuclei counter (DB+CNC) are presented. Both the EAA and DB+CNC can be modeled by the Fredholm Integral Equation of the First Kind. Thus, the data reduction methods for both the instruments are similar. The data reduction methods for both the instruments were analyzed with simulated and experimental data. For the EAA, the comparison between the different data reduction methods for the number distribution is excellent. The agreement between the number distribution retrieved by the three data reduction methods for the DB+CNC was found to be good. Finally, it was shown that for large number mean diameters, the response of the diffusion battery does not change significantly with a considerable change in the dispersion parameter, sigma(g). Thus, the resolution of sigma(g) with the diffusion batter is poor.

KEYWORDS: *Particle size distribution, *Monitors, *Air pollution, *Air pollution detection.

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PROGRESS REPORT

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"Research on Air Sampling Filter Media"

Data Reduction Techniques for Aerosol Size Distribution Measuring Instruments

By

Abde Kapadia

submitted by

Benjamin Y.H. Liu
Principal Investigator

Particle Technology Laboratory
Mechanical Engineering Department
University of Minnesota
Minneapolis, Minnesota

to

United States Environmental Protection Agency
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DATA REDUCTION TECHNIQUES FOR
AEROSOL SIZE DISTRIBUTION MEASURING INSTRUMENTS

A THESIS
SUBMITTED TO THE FACULTY OF THE GRADUATE SCHOOL
OF THE UNIVERSITY OF MINNESOTA

by

Abde Kapadia

IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY

June, 1980

i.a

ABSTRACT

Data reduction methods for the electrical aerosol analyzer (EAA) and the diffusion battery+condensation nuclei counter (DB+CNC) are presented. Both the EAA and DB+CNC can be modeled by the Fredholm Integral Equation of the First Kind. Thus, the data reduction methods for both the instruments are similar.

For the EAA, four data reduction methods are presented. First, an optimized EAA channel method is presented, in which the channel boundaries are chosen such that the response matrix can be approximated by an identity matrix. This method is applicable only for specialized applications of the EAA. The next two methods are based on two nonlinear algorithms (NLIA). When using the NLIA's, no assumption regarding the form of the aerosol size distribution must he made. Finally, a curve-fitting procedure based on a simplex minimization algorithm is investigated. This method gives the parameters of unimodal or bimodal log-normal distribution which best fits the data.

The two NLIA's and the curve-fitting procedure were also applied to the DB+CNC data reduction problem.

The data reduction methods for both the instruments were analyzed with simulated and experimental data. For the EAA, the comparison between the different data reduction methods for the number distribution is excellent. However, for the surface and volume distributions, the agreement was only satisfactory in the size range of 0.316 to 1.0 µm.

The agreement between the number distribution retrieved by the three data reduction methods for the DB+CNC was found to be good. Also, between different diffusion battery measurements for the same aerosol, the geometric number mean diameters (DGN) determined by the simplex minimization curve-fitting procedure agree satisfactorily. However, the agreement in the various estimates of the geometric standard deviation becomes unsatisfactory as the DGN increases. Finally, it was shown that for large number mean diameters, the response of the diffusion battery does not change significantly with a considerable change in the dispersion parameter, σ_g. Thus, the resolution of σ_g with the diffusion battery is poor.

sigma(g)

i.c

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NOMENCLATURE

C	Slip correction to Stokes' law
C'	Constant in power law aerosol size distribution function
C''	Constant in power law aerosol size distribution function
D	Diffusion coefficient of particle
D'	Equivalent diffusion coefficient
DGN	Number geometric mean diameter
DGN_1	Number geometric mean diameter for mode 1 (μm)
DGN_2	Number geometric mean diameter for mode 2 (μm)
DGS	Surface geometric mean diameter
DGS_1	Surface geometric mean diameter for mode 1 (μm)
DGS_2	Surface geometric mean diameter for mode 2 (μm)
DGV	Volume geometric mean diameter
DGV_1	Volume geometric mean diameter for mode 1 (μm)
DGV_2	Volume geometric mean diameter for mode 2 (μm)
D_i	Diffusion coefficient of the ith component in exhaustion method
D_o	Mean diffusion coefficient in method of Nolan and Scott
D_p	Particle diameter (μm)
D_{p_L}	Lower limit for particle diameter in power law aerosol size distribution
D_{p_u}	Upper limit for particle diameter in power law aerosol size distribution
$\overline{H}$	Matrix for Phillips-Twomey method
I_{o_j}	Total electrometer current for particles of diameter D_{p_j} with voltage on the central rod equal to zero (pA)

K_{ij}	Discretized form of kernel of the FIE, where subscript i refers to instrument variable v_i and subscript j, to particle diameter D_{p_j}
$K(v,D_p)$	Kernel of the FIE
N_I	Indicated number concentration by CNC
N_T	Total number concentration (part/cc)
N_0	True number concentration
$\overline{P}$	Centroid of a hyperplane of simplex
p^*	Simplex vertex obtained by reflection
p^{**}	Simplex vertex obtained by expansion
p^{***}	Simplex vertex obtained by contraction
$P(D_p)$	Penetration of particle size D_p
P_h	Vertex of a simplex which has the highest value of objective function
P_i	Fraction of the ith component in exhaustion method
P_j	Vertex of a simplex
R_{ij}	Response matrix of an instrument for particles of diameter D_{p_j} (for the EAA, subscript i refers to the ith channel, and for DB, i refers to the ith stage)
$S(D_p)$	Sensitivity of the instrument
$S_{N,T}$	Total number sensitivity of the EAA for a given log-normal aerosol size distribution ($pA/10^6$ part/cc)
SSRD	Sum of squares of relative differences
$S_{S,T}$	Total surface sensitivity of the EAA for a given log-normal aerosol size distribution ($pA/10^6\ \mu m^2/cc$)
S_T	Total surface concentration ($\mu m^2/cc$)
$S_{V,T}$	Total volume sensitivity of the EAA for a given log-normal aerosol size distribution ($pA/10^6\ \mu m^3/cc$)

S_j	Discretized form of instrument sensitivity $S(D_p)$ where subscript j refers to particle diameter D_{pj}
$S_{N,j}$	Monodisperse number sensitivity of the EAA for aerosol of diameter D_{pj} (pA/10^6 part/cc)
$S_{S,j}$	Monodisperse surface sensitivity of the EAA for aerosol of diameter D_{pj} (pA/10^6 μm^2/cc)
$S_{V,j}$	Monodisperse volume sensitivity of the EAA for aerosol of diameter D_{pj} (pA/10^6 μm^3/cc)
$S_{N,i}$	Polydisperse number sensitivity of the EAA for a log-normal aerosol size distribution for the ith channel (pA/part/cc)
$S_{S,i}$	Polydisperse surface sensitivity of the EAA for a log-normal aerosol size distribution for the ith channel (pA/μm^2/cc)
$S_{V,i}$	Polydisperse volume sensitivity of the EAA for a log-normal aerosol size distribution for the ith channel (pA/μm^3/cc)
V_T	Total volume concentration (μm^3/cc)
WLS	Weighted least squares
Z_p	Electrical mobility of particle (cm^2/volt-sec)
a_0	Constant in modified gamma aerosol size distribution
b_0	Constant in modified gamma aerosol size distribution
e	Elementary unit of charge (4.8 x 10^{-10} esu)
$f(D_p)$	Aerosol size distribution
f_j	Discretized value of aerosol size distribution $f(D_p)$, where subscript j refers to particle diameter D_{pj}
f_{n_1}	Number fraction in mode 1
f_{s_1}	Surface fraction in mode 1
f_{v_1}	Volume fraction in mode 1
g_i	Discretized value of measured response $g(v)$, where subscript i refers to instrument variable v_i
$g(v)$	Measured response of an instrument

h_{ij}	Element of the Hessian matrix
k	Constant in exhaustion method
$\overline{n}$	Total concentration at outlet in exhaustion method
$n(D_p)$	Number size distribution of the aerosol
n_i	Inlet concentration of ith component in exhaustion method
$\overline{n}_i$	Outlet concentration of ith component in exhaustion method
n_o	Total concentration at inlet in exhaustion method
n_p	Number of elementary charges
v	Instrument variable
v'	Viewing volume for CNC (cc)
v_i	Discretized value of instrument variable v
w_j	Weighting factor for quadrature formula
y	Parameter in exhaustion method
α	Constant for simplex minimization method
α_G	Constant in modified gamma aerosol size distribution
β	Constant for simplex minimization method
	Constant for simplex minimization method
$_i^p$	Parameter for NLIA's with subscript i corresponding to the ith measurement, and superscript p corresponding to the number of iterations
θ	Constant in modified gamma aerosol size distribution
θ'	Parameter for Phillips-Twomey method
Δ	Diffusion loss parameter
ΔI_i	Measured current difference in the ith channel for the EAA
$\Delta I_i'$	Calculated current difference in the ith channel for the EAA

$\Delta L_i'$	Calculated diffusion loss in the ith stage
ΔN_j	Number concentration of particles of diameter D_{pj} (part/cc)
ΔN_j^p	Number concentration of particles of diameter D_{pj} for the pth iteration of NLIA's
$\Delta \overline{N}$	Solution vector for Phillips-Twomey method
ΔS_j	Surface concentration of particles of diameter D_{pj} (μm^2/cc)
Δt_i	Measured diffusion loss in the ith stage
ΔV_j	Volume concentration of particles of diameter D_{pj} (μm^3/cc)
i	Discretization error for the ith measurement
i	Experimental error for the ith measurement
μ	Gas viscosity (poise)
	Power law exponent in power law size distribution function
D	Standard deviation of diffusion coefficient in method of Nolan and Scott
g	Geometric standard deviaion
g_1	Geometric standard deviation for mode 1
g_2	Geometric standard deviation for mode 2
i	Uncertainty of measured ΔI_i or ΔL_i

CHAPTER 1

INTRODUCTION

In recent years, considerable interest and concern has been expressed regarding submicron aerosols. Automobile exhaust and photochemical conversion of gaseous pollutants in the atmosphere usually produce aerosols in the submicron range. These aerosols affect human health, visibility, and climate. For example, submicron aerosols have a high penetration in the human respiratory tract. Also, the maximum scattering efficiency of light by a particle corresponds to the particle size of the same order as the wavelength of incident light.

As a first step in understanding the various phenomena associated with submicron aerosols, it is essential to know their size distribution and concentration. Various methods and instruments are available for measuring the concentration and size distribution of aerosols.

Since Aitken (1890-91) introduced the "Aitken Pocket Nuclei Counter", several manual and fully automatic condensation nuclei counters (CNC) have been developed. In the CNC, the aerosol is first saturated either with water vapor or alcohol in a chamber. Then supersaturation is produced in the chamber, either by adiabatic expansion or by cooling. In the supersaturated state, the vapor condenses on the particles, forming droplets. These are than measured optically either by light scattering or by light transmission principles. The CNC is used to measure the total particle concentration of submicron aerosols, but no information regarding the size distribution of the aerosol can be determined.

Particle size information for Aitken nuclei can be obtained by using a CNC to measure the penetration of submicron particles through a diffusion battery under steady flow conditions.

A diffusion battery consists of several stages. Each stage in turn consists of a bundle of channels of a definite geometric shape, generally the shapes being tubular or rectangular.

In order to measure the size distribution, the aerosol is drawn through various stages of the diffusion battery. Small particles in the stream diffuse towards the wall of the battery and deposit on the wall. The fraction of particles lost to the walls depends upon the particle size, the geometry of the diffusion battery, and flow rate. By means of a suitable data reduction procedure, the penetration values can then be converted to particle size distribution.

Since Whitby and Clark (1966) developed the first prototype of the electrical aerosol analyzer (EAA), the EAA has been used widely for measuring the size distribution of submicron aerosols, both in laboratory and in field measurements. The EAA requires about three to three and a half minutes to make a complete size distribution measurement. In this regard, it has considerable advantage over the CNC and diffusion battery methods, which may take anywhere between eight and fifteen minutes for a size distribution measurement.

In the EAA, the aerosol is first charged in a unipolar diffusion charger by positive ions. This charged aerosol then enters a mobility tube in which particles greater than a certain critical mobility are precipitated and the current due to the stream of remaining charged aerosol is measured. This is repeated for several values of critical mobility. The current measurements are finally converted to a size distribution using one of the several data reduction techniques available.

Nearly all instruments used for measuring the size distribution of aerosols use the indirect method of measurement, i.e., some size dependent property is measured, and from this, the size distribution is derived. For example, in the EAA, the current due to a charged stream of aerosol is measured as a function of the electrical mobility of the particles of a charged aerosol, and in the diffusion battery, the loss of particles due to diffusion is measured as a function of the length of the diffusion battery. It is desirable to have an instrument which has a one-to-one relationship between the measured property and the particle size, e.g., if the EAA were such an instrument, all the particles of the same size would have the same electrical mobility. In most real instruments, this is not the case, e.g., in the EAA, particles of the same size may have different electrical mobilities because particles of the same size may carry different quantities of electrical charge due to the statistical nature of the charging process.

To derive the aerosol size distribution from the experimental data, the response characteristics of the measurement instrument must be known. The response of both the CNC+diffusion battery method and the EAA can be modeled mathematically by the Fredholm Integral Equation of the First Kind (FIE), as given by Equation (1.1):

$$g(v) = \int S(D_p)\, K(v,D_p)\, f(D_p)\, d\, D_p \qquad (1.1)$$

where g(v) is a function of instrument variable v (the measured response), $S(D_p)$ (the sensitivity of the instrument) is a function of particle diameter D_p, $K(v,D_p)$ is the kernel of the integral equation which specifies the response function of the instrument, and $f(D_p)$ is the input "physical" aerosol size distribution. The value of g(v) is measured at several values of v, and $f(D_p)$ is the solution of the FIE which must be determined.

In principle, using a simple direct inversion technique after Equation (1.1) has been discretized, a solution representing $f(D_p)$ can be obtained. However, such a simple inversion procedure usually leads to a function which takes on negative values for some D_p and is also unrealistically oscillatory, bearing little resemblance to real aerosol size distribution. Thus, a more complicated technique involving a smoothing or an averaging method has to be applied to the data to retrieve the size distribution.

Twomey (1963, 1965, 1975) has suggested two such techniques. The first one involves a smoothing parameter, while the second is an iterative scheme for finding the solution to the FIE. Since no a priori knowledge of the value of the smoothing parameter is available, the first technique has not been widely used in data reduction problems encountered in aerosol science. The iterative technique has been adapted both for the CNC and diffusion battery system and the EAA by Knutson (1977). This is one of the techniques which has been used and evaluated in the present work for analyzing the data from these instruments. In addition, a new non-linear iterative algorithm is developed in the course of this thesis investigation and compared with Twomey's algorithm for analyzing the EAA and the diffusion battery data.

The third method used both for the diffusion battery and the EAA consists of finding a unimodal or bimodal log-normal distribution which best fits the data. A simplex minimization method developed by Nelder and Mead (1965) is adapted for use in finding the parameters of distribution which best fit the data. Sensitivities or correction factors are then calculated using the parameters of the optimized distribution. These correction factors, in conjunction with the measured data, are then used to obtain the final solution to the FIE.

The principal objective of this thesis is to make a systematic study of the several procedures which can be used for data reduction for aerosol size measurement instruments that can be modeled by Equation (1.1), to compare the various data reduction techniques using simulation and experimental data for the diffusion battery and the EAA and to identify their limitations.

To achieve this objective, first data reduction techniques were developed both for the diffusion battery system and the EAA. The utility of these data reduction methods was first determined by simulation studies. Once this was established, experimental data were taken for the EAA and the diffusion battery, both systems measuring the same aerosol. These data were then analyzed using their respective data reduction methods and the final solutions compared.

CHAPTER 2

ELECTRICAL AEROSOL ANALYZER: PRINCIPLE AND DATA REDUCTION

2.1 Introduction

This chapter is concerned with the electrical aerosol analyzer (EAA). Following a brief review of the history and operating principle of the EAA, the problem of data reduction for the EAA will then be treated.

The electrical aerosol analyzer has been used since 1971 to measure the size distribution and concentration of aerosol particles in the size range of 0.0032 μm to 1.0 μm. The EAA has been successfully used in both laboratory and field environments. The application of the EAA has been well summarized by Sem (1979).

The EAA is essentially an electrical aerosol mobility analyzer, first developed by Whitby and Clark (1966). An automated version of it called the Whitby Aerosol Analyzer (WAA), described by Whitby et al. (1972), was used for various field studies by the Particle Technology Laboratory (PTL) of the University of Minnesota, notably the 1969 Pasadena smog study (Hidy et al., 1972), Fort Collins, Colorado (Whitby et al., 1974), Denver, Colorado (Willeke et al., 1974) and the California ACHEX Program (Whitby et al., 1975; Sverdrup et al., 1975).

The WAA was rather large and heavy, and so a small portable version of the WAA, utilizing a different diffusion charger instead of the sonic jet diffusion charger, has been developed by B. Y. H. Liu and his students, principally D. Y. H. Pui. This instrument, called the EAA, has been used since the summer of 1971. A detailed account of the history and development of the EAA is given in a paper by Liu et al. (1979).

2.2 Operating Principle

The operating principle of the EAA involves first charging the aerosol, and then measuring the electrical mobility of the charged aerosol. When a charged particle is exposed to an electrical field, it experiences a force in the direction of the electrical field proportional to the product of the charge and the strength of the electrical field. When the particle starts moving in the direction of the force, it experiences a viscous drag which is given by Stokes' law. The electrical mobility, Z_p, is the terminal speed of the particle per unit electric field strength, and is given by Equation (2.1):

$$Z_p = \frac{n_p \; e \; C}{3 \pi \mu D_p (300)} \tag{2.1}$$

where Z_p is the electrical mobility (cm^2/volt-sec), n_p is the number of elementary charges on the particle, e is the elementary unit of charge (4.8^{-10} e.s.u.), C is the slip correction to Stokes' law, and μ is the gas viscosity in poise. The factor 300 is used to convert the electrostatic units to c.g.s. units. Figure 2.1 (Liu et al., 1979) shows the variation of the electrical mobility, Z_p vs. particle size for different ion concentrations, N_0, and charging time t (secs.) products. These curves are for the case when the particle charging mechanism is diffusion controlled, in contrast to the charging mechanism due to drift ions

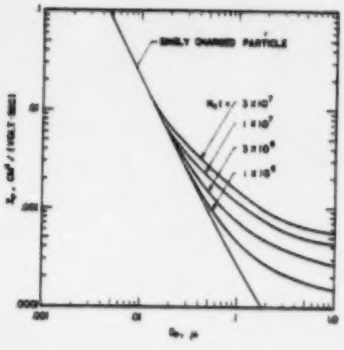

Figure 2.1 Theoretical relationship between mean electrical mobility and particle diameter for aerosols in unipolar diffusion charging (Liu et al., 1976).

in an externally applied field. In the EAA, a nominal value of 1.0 x 10^7 ions sec/cc is used for N_0t. It can be seen that the mean electrical mobility of the particles is a monotonic decreasing function of particle size up to about 1.0 μm. Thus, if the electrical mobility of a diffusion charged aerosol is known, the particle size can be easily determined for particles of diameter less than 1.0 μm.

2.3 Description of EAA

Figure 2.2 (Liu and Pui, 1975) gives the schematic of the EAA. The EAA consists primarily of three components: (a) the diffusion charger, (b) the mobility tube, and (c) the electrometer.

Four liters per minute of aerosol together with one liter per minute of charger sheath air enters the diffusion charger. In the charger, the particles are exposed to a stream of positively charged ions, produced by corona discharge from a wire in the center of the charger. The aerosol particles are thus charged by diffusion of the ions to the particles. The charged aerosol then enters the mobility tube in the form of an outer annular sheath, surrounding 45 lpm of clean sheath air which flows next to the inner cylinder. When a negative voltage is applied to the inner cylinder, all particles greater than a certain critical mobility are precipitated on the inner cylinder, while the rest of the charged particles flow to the electrometer. In the electrometer current sensor, the particles are then collected on an absolute filter and the charge due to the particles is measured as a current by the electrometer. This current is converted to a voltage and displayed on a digital voltmeter.

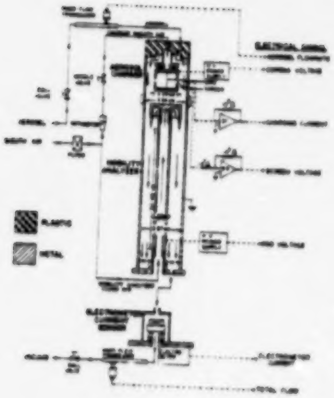
PLASTIC
METAL

Figure 2.2 Schematic diagram of the electrical aerosol analyzer (Liu and Pui, 1975).

In the normal operating mode of the EAA, the collector rod voltage is varied from -20 volts to -10,600 volts in 11 steps. As the absolute value of the voltage is increased, particles with lower mobilities are precipitated on the central rod, and the current in the electrometer decreases. By subtracting the currents from adjacent voltage steps, the current due to particles within corresponding mobilities is determined. Knowing the particle electrical mobility and the instrument sensitivity as a function of particle size, the number size distribution and the total particle concentration can be determined.

The EAA covers the particle size range from 0.0032 μm to 1.0 μm. The lower size limit of the instrument is set by the high loss of the smaller size particles in the charger. Also, a very small fraction of the smaller particles get charged in the diffusion charger, and thus the sensitivity of the EAA also becomes very small.

From Figure 2.1, it can be seen that near 1.0 μm, for an N_0t product of 1.0×10^7 ion sec/cc, the electrical mobility vs. particle size curve becomes almost flat. This decreasing dependency of the mobility with the particle size establishes the upper limit of the instrument. Moreover, a large fraction of particles greater than 1.0 μm are also lost in the flow lines within the instrument. A more detailed explanation of the operating principle and the components of the EAA is given by Liu et al. (1979).

2.4 Calibration of EAA

As mentioned previously, in order to get the size distribution of the aerosol and total particle concentration, the variation of the mobility of the diffusion charged aerosol with particle size and the relationship between the sensitivity of the instrument and particle size must be established.

Figure 2.1 gives the theoretical relationship between the mean electrical mobility and particle size for particles charged by a diffusion controlled mechanism for different N_0t products. These theoretical calculations cannot be directly applied to the EAA, as the space charge effect has been neglected in the theory. Moreover, there is a weak electric field (approximately 55 volts/cm) in the charging region, and therefore, the actual charging rate may differ from that predicted by a pure diffusion charging theory. Also, the exact mobility of the ions in the charging region is difficult to determine, and therefore, the ion concentration can be determined only approximately. Thus, the charging characteristics of the charger in the EAA must be determined experimentally.

In order to calibrate the EAA, Liu and Pui applied known concentrations of monodisperse aerosols of known sizes to the EAA and determined its response. A more detailed account of the generation of monodisperse aerosols used in the calibration of the EAA is given in Pui and Liu (1979).

The instrument sensitivity was determined for several sizes of monodisperse aerosols using Equation (2.2):

$$\dot{S}_{N,j} = \frac{I_{e_j} \times 10^6}{\Delta N_j} \qquad (2.2)$$

where $S_{n,j}$ is the number sensitivity of the EAA for aerosol of diameter D_{pj} (in pA/1.0x10^6 part/cc), I_{pj} is the electrometer current in pA, and ΔN_j is the number concentration of particles of diameter D_{pj} (in particles/cc). The electrometer current was then measured as a function of the collector rod voltage. This gave a family of voltage vs. current curves, which provided the data for characterizing the diffusion aerosol charger and the overall performance of the EAA.

Figure 2.3 (from Liu and Pui, 1975) gives the sensitivity of the instrument as a function of particle size. The sensitivity is a strong function of the particle size, varying from 400 pA/(1.0x10^6 part/cc) at 1.0 μm, to a low of 0.056 pA/(1.0x10^6 part/cc) for 0.0072 μm. For sensitivity below 0.0072 μm, Liu and Pui extrapolated the data, as no suitable monodisperse aerosols of sizes below 0.0072 μm could be produced.

Equation (2.3) (from Sverdrup, 1977) gives the equations to the original data points of Liu and Pui obtained by regression:

$$\dot{S}_{n,j} = 2.351 \times 10^{12} \times D_{pj}^{6.262} \qquad 0.0032\ \mu m \le D_{pj} < 0.0125\ \mu m$$

$$= 426.4 \times D_{pj}^{1.156} \qquad 0.0125\ \mu m \le D_{pj} \le 2.05\ \mu m \qquad (2.3)$$

where D_{pj} is the particle diameter in microns. Very often the EAA is used as an aerosol detector. In this mode of operation, the current from the EAA is measured with the voltage on the central rod set to zero or -19 volts, which corresponds to the voltage for Step 1 in the EAA.

In most cases, the EAA is used to measure a size distribution which can be approximated well by a log-normal distribution. In such

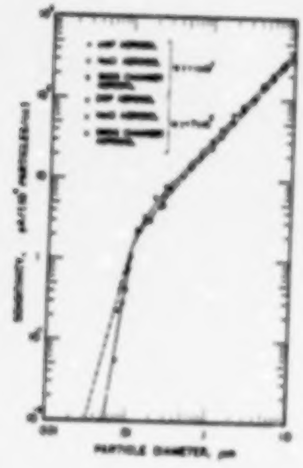

Figure 2.3 Sensitivity of the EAA as a function of particle diameter (Liu and Pui, 1975).

applications of the EAA, the total number, surface, and volume sensitivities of the instrument would be useful. The theoretical sensitivities of the instrument can be calculated by integrating the sensitivity function given in Equation (2.3) over the aerosol size distribution. Equations (2.4), (2.5), and (2.6) give the expressions for the total sensitivities. Figures 2.4, 2.5, and 2.6 show the variation of the total sensitivity with the geometric means for various geometric standard deviations.

$$S_{N,T} = \int S_N(D_p)\, f_N\,(D_p,\, DGN,\, \sigma_g)\, d\ln D_p \qquad (2.4)$$

$$S_{S,T} = \int \frac{S_N(D_p)}{\pi(D_p)^2}\, f_S\,(D_p,\, DGS,\, \sigma_g)\, d\ln D_p \qquad (2.5)$$

$$S_{V,T} = \int \frac{S_N(D_p)}{\frac{\pi}{6}(D_p)^3}\, f_V\,(D_p,\, DGV,\, \sigma_g)\, d\ln D_p \qquad (2.6)$$

where $S_{N,T}$, $S_{S,T}$, and $S_{V,T}$ are the total sensitivities in pA/(1.0x10^6 part/cc), pA/(1.0x10^6 μm^2/cc), and pA/(1.0x10^6 μm^3/cc), respectively, and

$$f_K\,(D_p,\, DGK,\, \sigma_g) = \frac{1}{\sqrt{2\pi}\,\ln\sigma_g} \exp - \frac{1}{2}\, \frac{\ln D_p/DGK}{\ln\sigma_g}^2$$

K = N for number distribution

= S for surface distribution

= V for volume distribution

is the log-normal distribution where DGN, DGS, and DGV are the number, surface, and volume geometric mean diameters, respectively, and σ_g is the geometric standard deviation. Using Figures 2.4 to 2.6, the total number, surface, and volume concentration can be easily

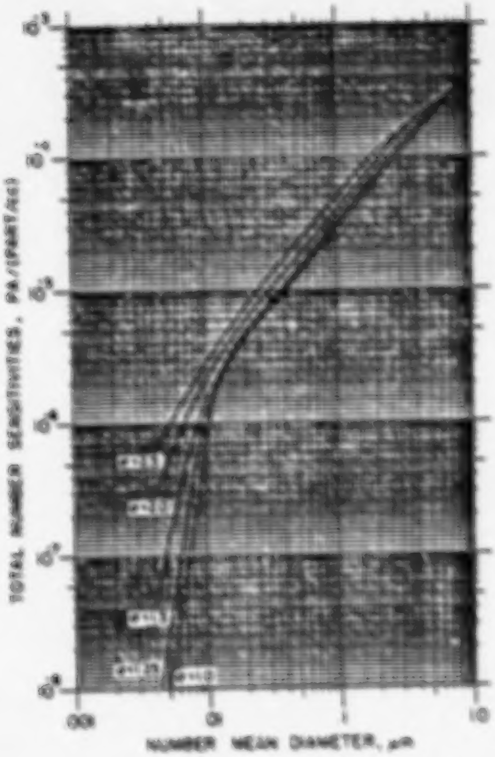

Figure 2.4 Total number sensitivities for log-normal aerosol size distributions with various number mean diameters and geometric standard deviations.

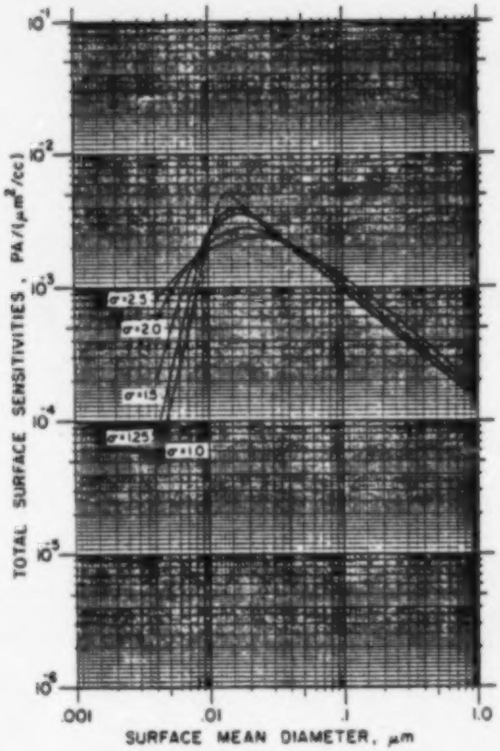

Figure 2.5 Total surface sensitivities for log-normal aerosol size distributions with various surface mean diameters and geometric standard deviations.

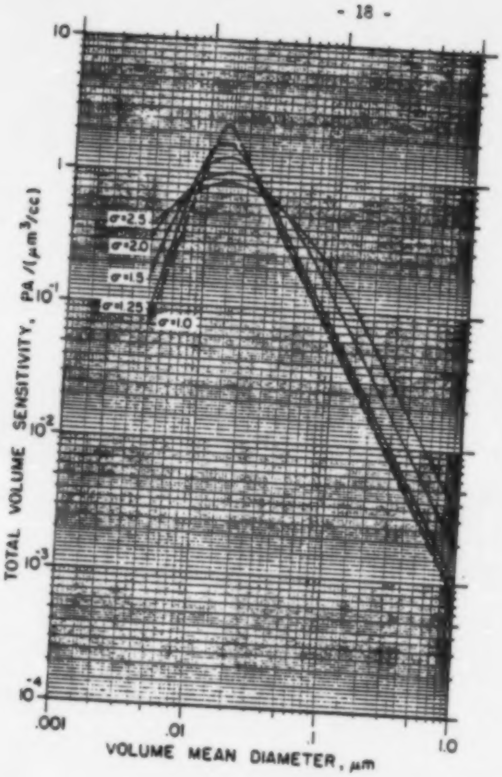

Figure 2.6 Total volume sensitivities for log-normal aerosol size distributions with various volume mean diameters and geometric standard deviations.

determined if the geometric standard deviation and geometric mean are known.

Figure 2.7 (from Liu and Pui, 1975) shows the variation in the electrometer current with voltage for various monodisperse aerosols. The electrometer current has been made dimensionless by dividing the measured current with the maximum measured electrometer current. Several important features of these curves were noted by Liu and Pui. First, it can be seen that the curves of aerosols of diameter less than 0.018 μm, and greater than 0.18 μm, are straight lines, while the current vs. voltage curves for particles between 0.018 μm and 0.18 μm have discontinuities in them. The curve for 0.018 μm particles, as mentioned previously, is a straight line and has a very large negative slope. This has been explained by Liu and Pui by the fact that the particles of 0.018 μm, which were generated by the electrical classification method, were highly monodisperse and that all the charged particles carried a single unit of charge. On the other hand, the curves for 0.0125 μm and 0.0105 μm are not as steep. These aerosols were generated in a smog chamber and had a much larger geometric standard deviation than those aerosols generated by the electrostatic classification method.

The current vs. voltage relationships for monodisperse aerosols of sizes ranging from 0.024 μm to 0.13 μm show discontinuities. These discontinuities are explained by the multiple charging which takes place in the diffusion charger of the EAA, e.g., for particles of size 0.024 μm. About 5% of the particles are doubly charged, while the rest of the particles are singly charged. As the particle size increases, the ratio

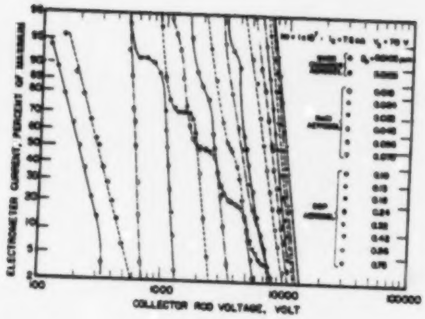

Figure 2.7 Voltage vs. current curves for EAA for an N_0t product of 1 x 10^7 (ions/cc)sec (Liu and Pui, 1975).

of the multiply charged particles to singly charged particles increases. In fact, for 0.0056 μm particles, Liu and Pui show that about 7% of the particles are triply charged, 30% doubly charged, and the rest singly charged.

For particle sizes larger than 0.13 μm, the discontinuities in the curves again disappear. This can be explained by the fact that these particles are so large that they acquire numerous elementary charges, and thus the discrete effect of these charges becomes continuous.

It can also be seen that as the particle size increases, the spacing between the curves decreases, though the logarithmic size increments for the particle diameter are the same. This is because the electrical mobility changes rapidly with diameter for small particles, but for large particles, the change in mobility is not as great, as can be seen from Figure 2.1.

Appendix A gives the data of Figure 2.7 in tabular form.

2.5 Data Reduction

2.5.1 Statement of the Problem

Nearly all the instruments used for measuring size distributions of aerosols use the indirect method of measurement, i.e., some size-dependent property is measured, and from these measurements, the size distribution is then deduced, e.g., in the EAA, the current as detected by the electrometer is measured as a function of the voltage applied to the central rod.

For an ideal instrument, there is a one-to-one relationship between the measured property and the particle size. However, in particle size measuring instruments, this is rarely the case, e.g., in the EAA, particles of the same size can have different mobilities due to multiple charging effects of the diffusion charger, and therefore, particles of different sizes can have the appropriate critical mobility and thus are precipitated on the central rod when a voltage is applied to it. Thus, the inversion of the current vs. voltage curve to the size distribution becomes more complicated.

Mathematically, instruments like the EAA can generally be modeled by Equation (2.7):

$$g(v) = \int_0^\infty S(D_p)\ K(v,D_p)\ f(D_p)\ dD_p \qquad (2.7)$$

where g(v) is the response of the instrument (e.g., in the EAA, the current is the response which is measured as a function of voltage), $S(D_p)$ is the sensitivity of the instrument, $K(v,D_p)$ is the kernel or the response function of the instrument (e.g., in the EAA, the fraction of current as a function of voltage and particle size), and $f(D_p)$ is the function to be determined (e.g., in the EAA, the particle size distribution). In literature, equations such as (2.7) are referred to as the Fredholm Integral Equation of the first kind (FIE). The functions $K(v,D_p)$ and g(v) are assumed to be continuous or, at least, bounded and piecewise continuous. The FIE does not have a solution for every g(v), and when it is solvable, there may be no unique solution. Even if a unique solution exists, it may be highly unstable, in the sense that a small change in g(v) may produce large changes in $f(D_p)$.

In an experimental situation, either one or both of the variables, D_p or v, could be discrete or continuous. For example, in the EAA, v is a discrete variable while D_p is continuous. However, for calculation purposes, D_p is generally also discretized. Equation (2.7) can then be replaced by a set of linear equations given by Equation (2.8):

$$g_i = \sum_{j=1}^{J} S_j K_{ij} \Delta N_j \qquad i=1,\ldots I \tag{2.8}$$

The discretization process can be explained in the following manner (Rust and Burrus, 1972). If only discrete values of v are taken into consideration, Equation (2.7) can be written as

$$g(v_i) = \int_0^\infty S(D_p)\, K(v_i,D_p)\, f(D_p)\, dD_p \tag{2.9}$$

where v_i are the discretized values of the voltages corresponding to the EAA channels. There is no approximation or discretization error involved in writing Equation (2.9). To discretize the variable D_p, the integral is replaced by a quadrature formula, as in Equation (2.10):

$$g_i = \sum_{j=1}^{J} w_j S_j K_{ij} f_j + \delta_i \qquad i=1,\ldots I \tag{2.10}$$

where w_j is the weight depending upon the quadrature formula used. The use of this quadrature formula introduces the discretization error, δ_i, and the magnitude of this error depends upon the value of J and the quadrature formula used. By solving the system of linear equations given in Equation (2.8), negative values of f_j are obtained, which is physically unrealistic. These physically unrealistic solutions are obtained not because of the discretization errors, but because of the inherent ill-conditioned nature of the FIE itself, i.e., an infinitesimal change in either g_i, S_j, or K_{ij} produces finite changes in f_j, or that the solution to Equation (2.10) is extremely sensitive to the coefficient matrix and the data vector. This has been borne out by the fact that even though the error due to quadrature is made small by considering a larger value of J in Equation (2.8), negative values of f_j are still obtained and sometimes greater ill-conditioning results. Similar results have been reported by Rust and Burrus (1972) for several different FIE's.

To take into account the errors due to experimentation, the set of linear equations given in Equation (2.8) are written as

$$g_i = \sum_{j=1}^{J} S_j K_{ij} \Delta N_j + \epsilon_i \qquad i=1,\ldots I \tag{2.11}$$

where $\Delta N_j = w_j f_j$ and $_i$ is a random normal deviate with mean zero and an appropriate sigma which takes the experimental repeatability into consideration.

2.5.2 Methods for Solving the FIE

A numerical method for solving the set of equations similar to Equation (2.11) was presented by Phillips (1962) and later modified by Twomey (1963). In the Phillips-Twomey method, the smoothest acceptable solution to Equation (2.11) is found by minimizing the second difference expression

$$\sum_{j=1}^{J} (\Delta N_{j+1} - 2\Delta N_j + \Delta N_{j-1})^2$$

with Equation (2.11) as the auxiliary conditions, and

$$\sum_{i=1}^{I} \epsilon_i^2 = \text{constant}$$

The quantity to be minimized is a weighted sum of the second differences and the sum of squares of the residuals, as given by Equation (2.12):

$$Z = \sum_{j=1}^{J} (\Delta N_{j+1} - 2\Delta N_j + \Delta N_{j-1})^2 + \frac{1}{\alpha} \left(\sum_{i=1}^{I} \epsilon_i^2 \right) \qquad (2.12)$$

The solution of the system of linear equations is given by

$$\Delta \overline{N} = (\overline{R}^* \overline{R} + \gamma_0' \, \overline{H})^{-1} \, \overline{R}^* \, \overline{G} \qquad (2.13)$$

where $\overline{R}$ is a matrix whose element r_{ij} is $S_j K_{ij}$, $\overline{R}^*$ is the transpose of $\overline{R}$, γ_0' is an undetermined constant, $\overline{H}$ is a matrix such that $| \Delta\overline{N}^* \, \overline{H} \, \Delta\overline{N} |$ is a measure of the second difference of the number size distribution in finite difference form, and is given by Equation (2.14), G is the vector of measurements, $g(v_i)$, and $\overline{\Delta N}$ is the solution vector.

$$H = \begin{vmatrix} 1 & -2 & 1 & 0 & 0 & 0 & 0 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ -2 & 5 & 4 & 1 & 0 & 0 & 0 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 1 & -4 & 6 & -4 & 1 & 0 & 0 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 0 & 1 & -4 & 6 & -4 & 1 & 0 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 0 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & 1 & -4 & 5 & -2 \\ 0 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & 1 & -2 & 1 \end{vmatrix} \qquad (2.14)$$

If a very large value is given to γ_0', then there is excessive smoothing and important characteristics of the function to be retrieved (ΔN_j, $j=1,\ldots J$) are lost, while giving a small value to γ_0' ($=0$), a highly oscillating solution of ΔN_j is obtained. Thus, it is very essential to find an appropriate or a reasonable value of γ_0' if the Phillips-Twomey method is to be used for data reduction in aerosol size measuring instruments. There is no a priori method for finding a reasonable value of γ_o' for aerosol size distributions, and thus, this method has not been used extensively for data reduction in aerosol science.

Twomey (1975) presented an iterative non-linear algorithm for solving equations similar to (2.11). The procedure is as follows: if γ_i^{p-1} denotes the ratio of the observed g_i to

$$\sum_{j=1}^{J} S_j K_{ij} \Delta N_j^{p-1}$$

then the pth iteration is given by

$$\Delta N_j^p = [1 + (\gamma_i^{p-1} - 1) K_{ij}] \Delta N_j^{p-1} \qquad \begin{array}{l} j=1,\ldots J \\ i=1,\ldots I \end{array} \tag{2.15}$$

where

$$\gamma_i^{p-1} = g_i / \sum_{j=1}^{J} S_j K_{ij} \Delta N_j^{p-1}$$

To start the iterative procedure, an initial estimate of the number distribution, i.e. ΔN_j^o, $j=1,\ldots J$, is made. Based on the initial estimate, γ_1^o is first calculated, and a new set of ΔN_j^o, $j=1,\ldots J$ are determined. It should be noted here that the superscript p, which represents the number of iterations, still remains zero. Based on the new ΔN_j^o,$j=1,\ldots J$, γ_2^o and then new ΔN_j^o, $j=1,\ldots J$ are computed. This is repeated for $i=1,\ldots I$. After the $i=I$ update, p is incremented by unity. This procedure is repeated several times until a convergence criteria is met.

The implicit assumptions in this algorithm are that if the initial estimates of ΔN_j (ΔN_j^1) are greater than zero, then ΔN_j^p will always remain positive if K_{ij} is positive and <1.0. For aerosol size measuring instruments, K_{ij}'s are always >0 and <1.0, therefore, $\Delta N_j^p > 0.0$. In this algorithm, if ${}_i^{p-1}$ is greater than 1.0, then the term in the box brackets will be greater than 1.0, and thus, ΔN_j^p will be greater than ΔN_j^{p-1}. This will tend to decrease Y_i^p. On the other hand, if Y_i^{p-1} is less than unity, the term in the box brackets will be less than unity but greater than 0, as $K_{ij} > 0.0$. Thus, ΔN_j^p will be less than ΔN_j^{p-1}, which will tend to increase Y_i^p. If Y_i^{p-1} is unity, ΔN_j^p remains unchanged, and the iterative procedure will have finally converged. This non-linear algorithm was first applied to the EAA and the diffusion battery by Knutson (1977).

In Twomey's NLIA, for one complete iteration, I updates of ΔN_j, $j=1,\ldots J$ are required, with each update taking into account the discrepancy between the intermediate solution and one data point. The complexity of the iterative procedure and the number of computational operations can be reduced if, for each iteration, only one update of ΔN_j, $j=1,\ldots J$ is required. The correction factor for such an update should, obviously, take into account the discrepancy between the intermediate solution and the whole data set. The second non-linear iterative algorithm [NLIA(II)], developed during the course of this investigation, given by Equation (2.16), has the above-mentioned properties.

$$\Delta N_j^p = \Delta N_j^{p-1} \times \left(\sum_{i=1}^{I} K_{ij}\, Y_i^{p-1} \right) \quad \sum_{i=1}^{I} K_{ij} \qquad j=1,\ldots J \qquad (2.16)$$

In this algorithm, Y_i^0, $i=1,\ldots I$ are first calculated based on the initial estimate, ΔN_j^0, $j=1,\ldots J$. Each ΔN_j is then updated by multiplying the present value of ΔN_j with a weighted sum of the Y_i's, the weighting factors being the element of the response matrix corresponding to the particular diameter and the data point. Like Twomey's algorithm, this algorithm also gives non-negative ΔN_j's, if the initial ΔN_j's are >0.0. In NLIA(II), if all the γ_i^{p-1}'s are greater than unity, the quantity in curly brackets, the correction factor, will be greater than 1.0. It should be noted here that for the EAA and DM+CMC in the normal operating mode,

$$\sum_{i=1}^{I} K_{ij} = 1.0$$

and thus, Equation (2.16) can be written as

$$\Delta N_j^p = \Delta N_j^{p-1} \times \mathop{}_{i=1}^{I} K_{ij}\, \gamma_i^{p-1} \qquad j = 1,\ldots,J$$

Thus, ΔN_j^p will be greater than ΔN_j^{p-1}, which will tend to decrease γ_i^p. Similarly, if γ_i^{p-1}'s are less than unity, the ΔN_j^p will be less than ΔN_j^{p-1}. If some of the γ_i^{p-1}'s are greater than unity and others less than 1.0, the value of the correction factor will then be a weighted average of all the γ_i^{p-1}, with K_{ij} being the weighting factors. Also, for all γ_i^{p-1} equal to unity, the correction factor will be equal to 1.0, and ΔN_j^p will remain unchanged and the iterative procedure will have converged.

This algorithm has the advantage that it needs approximately two-thirds the computing operations compared to Twomey's algorithm for a complete iteration.

2.6 EAA Data Reduction Methods

2.6.1 Review of Previous Work

Liu and Pui (1975), in their calibration work for the EAA, determined the monodisperse sensitivities for the EAA for several particle sizes. In that paper, they also present an 11x11 response matrix for the EAA (Table 2.1). To calculate the aerosol size distribution, they suggest that first the difference in the measured electrometer current between adjacent voltage steps be determined. The particle concentration within each channel is then given by the ratio of the current difference and the sensitivity of the particles, corresponding to the geometric midpoint of the channel. From the number distributions, the surface and volume distribution are determined by multiplying the ΔN_j with $\pi(D_{pi})^2$ and $\frac{\pi}{6}(D_{pi})^3$, where D_{pi} is the geometric midpoint of the channel.

There are two implicit assumptions made when the EAA data is reduced in the manner suggested by Liu and Pui. First of all, this data reduction procedure does not make full use of the calibration matrix determined by them, but assumes an identity matrix as the calibration matrix.

On examining the response matrix in Table 2.1, it can be noted that for 0.042 μm diameter monodisperse aerosol, only 7% of the current appears in the fifth channel, while the remaining current due to the 0.042 μm particles contributes to the ΔI_i in the 4th and the 6th channels. Thus, the monodisperse sensitivity method underestimates the concentration of particles in the 5th channel, while overestimating the concentration in the adjacent channels. Thus, the aerosol size distribution obtained by the monodisperse sensitivity method can have considerable error; however, the estimate of the total number concentration may be more accurate.

Table 2.1 Response of the electrical aerosol analyzer to monodisperse aerosols for $N_t = 1 \times 10^7$ (ions/cc)sec (4 channels per decade) (Liu and Pui, 1975)

			Particle diameter(μm)	0.0042	0.0075	0.013	0.024	0.042	0.075	0.13	0.24	0.42	0.75
			Sensitivity pA/ 10^6 part/cc		0.105	2.40	6.00	11.5	22.5	41.5	81.0	150	285
Channel	Channel Midpoint (μm)	Channel Boundary (μm)	Collector rod voltage (volts)										
		0.0032	19										
1	0.0042				0	0	0	0	0	0	0	0	0
		0.0056	55										
2	0.0075				1	0	0	0	0	0	0	0	0
		0.0100	186										
3	0.0133				0	1	0.08	0	0	0	0	0	0
		0.0178	588										
4	0.0237				0	0	0.92	0.49	0	0	0	0	0
		0.0316	1070										
5	0.0422				0	0	0	0.07	0.14	0	0	0	0
		0.0562	2600										
6	0.075				0	0	0	0.44	0.65	0.42	0	0	0
		0.100	4440										
7	0.133				0	0	0	0	0.17	0.49	0.22	0	0
		0.178	6600										
8	0.237				0	0	0	0	0.02	0.09	0.48	0.31	0.11
		0.316	8380										
9	0.422				0	0	0	0	0.02	0	0.20	0.36	0.28
		0.562	9600										
10	0.750				0	0	0	0	0	0	0.06	0.18	0.25
		1.000	10600 (>10600)								(0.04)	(0.15)	(0.36)

Secondly, it approximates the aerosol size distribution by ten monodisperse aerosols whose diameters are the geometric midpoints of the EAA channels. As the EAA measures the continuous size distribution by making several discrete measurements, some assumption regarding the aerosol size distribution cannot be avoided. As the channel widths of the EAA are reasonably small, the second assumption may not have an appreciable effect on the final distribution.

To overcome the shortcomings of the monodisperse sensitivity method of Liu and Pui (1975), Whitby and Cantrell (1979) first formed a modified 11x11 response matrix. Using this matrix and a model of the ambient aerosol, they computed a set of constants for the ten EAA channels. Using these constants instead of the monodisperse sensitivities, they reduced the EAA data from field measurements. This method gives adequate results for aerosols which are similar to the ambient aerosol model. However, if the aerosol to be measured is significantly different from the ambient aerosol model, the results are not satisfactory, and one has to restart the process by calculating a new set of constants.

Liu et al. (1979) took this method one step further by introducing an iterative procedure for determining the constants, which they refer to as polydisperse sensitivities. Liu et al. (1979) present a set of tables of polydisperse sensitivities for various unimodal log-normal distributions with several geometric mean diameters and geometric standard deviations. Using the monodisperse sensitivities of Liu and Pui, a first approximation of the number size distribution is determined.

The geometric number mean diameter and the geometric standard deviation are determined for the approximate number size distribution. Then, using the polydisperse sensitivities from the table for the appropriate number size distribution, a second approximate solution for the number size distribution is calculated. This procedure is repeated until the geometric mean diameter and the geometric standard deviations from two consecutive iterations do not differ appreciably.

This method gives satisfactory results for unimodal distributions, but is rather tedious to use.

2.6.2 Present Work

In the following paragraphs, four new EAA data reduction methods, together with their limitations, are discussed. First, an optimized EAA channel method is presented in which the channel boundaries are chosen such that the response matrix can be approximated by an identity matrix. The next two methods are based on the two non-linear iterative algorithms discussed previously. When using the NLIA's, no assumption regarding the form of the aerosol size distribution must be made. Finally, a curve fitting procedure based on Nelder and Mead's (1965) simplex minimization algorithm is investigated. This method gives the parameters of a unimodal or a bimodal log-normal distribution which best fits the data.

2.6.2.1 Optimized EAA Channel Method

The monodisperse sensitivity method of Liu and Pui (1975) is simple and fairly easy to use. It can give satisfactory results in the channels of the EAA are chosen so that the response matrix can be approximated by an identity matrix. By choosing appropriate voltages for the central rod in the EAA, the response matrix can be made such that the diagonal elements are nearly unity, and the off-diagonal elements, negligible. This was accomplished by replotting the data of Figure 2.7, as shown in Figure 2.8. The abscissa in Figure 2.8 is the particle diameter and the ordinate is the central rod voltage of the EAA. The 2% and 98% curves correspond to the voltage vs. diameter relationship for the corresponding percentage of the current. For particle size less than 0.018 µm, the curves are parallel to each other, with a relatively small spacing between them. As the particle size increases, the curves are no longer parallel, and the distance between them increases. This is due to the multiple charging effect of the diffusion charger for larger particles. To determine the optimum channel boundaries and width, a "staircase curve", as shown in Figure 2.8, was constructed. The midpoint of the new channels of the EAA are determined by the vertical lines of the staircase curve. To determine the bounds of the channels, the voltages corresponding to the horizontal lines are first determined. Taking these voltages as the median collector rod voltages, the boundaries of the channels can be determined from Figure 2.9 (Liu and Pui, 1975).

Table 2.2 gives the response matrix, the midpoint diameter, the bounds of the new channels, the new channel widths, and the number sensitivities for the staircase curve shown in Figure 2.8. Other staircase curves can also be constructed and the corresponding values determined. On examining the $\Delta \log D_p$ column in Table 2.2, it can be seen that the channel widths for the first five channels are much smaller than 0.25, which is the channel width in the EAA at present. Thus, the EAA is

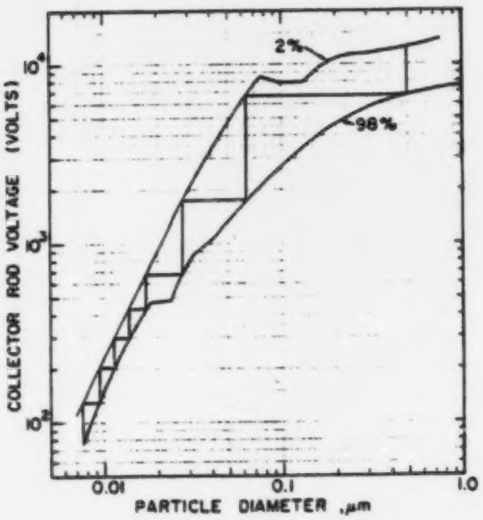

Figure 2.8 Central rod voltage vs. particle diameter relationships for 2% and 98% of the total current.

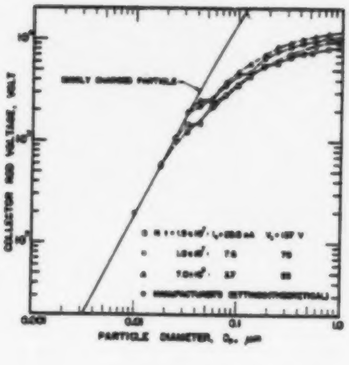

Figure 2.9 Median collector rod voltage vs. particle diameter for the EAA (Liu and Pui, 1975).

Table 2.2 Response of the electrical aerosol analyzer to monodisperse aerosols for $N_t = 1 \times 10^7$ (ions/cc)sec (optimized channels)

				Particle diameter (μm)	0.0075	0.0093	0.0112	0.0138	0.0170	0.0275	0.0622	0.500
				Sensitivity pA/ 10^6 part/cc	0.105	0.5	1.5	2.6	3.7	7.1	18	185
Channel	Channel Midpoint (μm)	Channel Boundary (μm)	$\Delta \log D_p$	Collector rod voltage (volts)								
		0.0063		75								
1	0.0075		0.12		0.98	0.02	0	0	0	0	0	0
		0.0083		130								
2	0.0093		0.10		0.02	0.96	0.02	0	0	0	0	0
		0.0105		205								
3	0.0112		0.083		0	0.02	0.96	0.02	0	0	0	0
		0.0127		300								
4	0.0138		0.078		0	0	0.02	0.96	0.02	0	0	0
		0.0152		435								
5	0.017		0.097		0	0	0	0.02	0.96	0.02	0	0
		0.019		680								
6	0.0275		0.213		0	0	0	0	0.02	0.96	0.02	0
		0.031		1750								
7	0.0622		0.799		0	0	0	0	0	0.02	0.96	0.02
		0.195		6800								
8	0.500		0.709		0	0	0	0	0	0	0.02	0.98
		1.000		12500								

capable of measuring much finer resolution in the size range up to 0.019 μm; however, the last two channels are much wider than 0.25. For particles in the size range of the last two channels, the resolution capability of the EAA is significantly poorer than for particles smaller than 0.018 μm in diameter. This is a direct implication of the diminishing slope of the electrical mobility vs. particle size curve of Figure 2.1 as the particle diameter increases.

In the optimized channel method, as in the method of Liu and Pui, the assumption of approximating the aerosol size distribution by a few monodisperse aerosols is implied. For the first five channels, as the channel width is reasonably narrow, this assumption will not have an appreciable effect on the reduced data; however, for the last two channels, the reduced data will not be satisfactory. Thus, the optimized channels of the EAA give useful information only in the size range from 0.0075 μm to 0.19 μm, and should be used only for specialized applications of the EAA.

2.6.2.2 NLIA's Applied to the EAA

As mentioned in the earlier paragraph, the NLIA can be used to solve the discretized form of FIE for the EAA, the set of simultaneous equations given by:

$$\Delta I_i = 10^6 \times \sum_{j=1}^{45} \hat{S}_{N,j} R_{ij} \Delta N_j + \epsilon_i \qquad i=1,\ldots 11 \qquad (2.17)$$

where ΔI_i is the measured current difference between two adjacent channels in pA; $\hat{S}_{N,j}$ is the number sensitivity of the instrument for the jth aerosol size particle in pA/1.0x10^6 particles/cc, as given by

Equation (2.2); R_{ij} is the element of the 11x45 response matrix for the EAA, as given by Liu et al. (1979); ΔN_j is the particle concentration of the jth size; and ϵ_i is the error associated with each measured ΔI_i. Equation (2.15), when applied to (2.17), becomes:

$$\Delta N_j^{p} = [1 + (\gamma_i^{p-1} - 1)\, R_{ij}]\, \Delta N_j^{p-1}$$

$$\gamma_i^{p-1} = \Delta I_i / \sum_{j=1}^{45} \hat{S}_{Nj}\, R_{ij}\, \Delta N_j^{p-1} \qquad \begin{matrix} j=1,\ldots 45 \\ i=1,\ldots 11 \end{matrix} \qquad (2.18)$$

In this technique, the logarithmic particle diameter scale was first divided into 16 equal divisions per decade. This procedure gave 45 divisions in the range between 0.0032 μm and 2.347 μm. The 45 divisions correspond to the subscript j in Equation (2.18) and the subscript i for $i=1,\ldots 10$ correspond to the ten measured current differences, while $i=11$ corresponds to the residual current in the 11th channel measurement.

In order to start the iterative procedure using Equation (2.18), first a starting solution is required. Knutson (1977) used a starting solution given by:

$$\Delta N_j^{1} = \sum_{i=1}^{11} \Delta I_i \quad \hat{S}_{N,j} \times 45 \qquad j=1,\ldots 45 \qquad (2.19)$$

Equation (2.19) assumes a uniform current distribution over the particle size range of the EAA. At a first glance, Equation (2.19) appears to estimate a very large particle concentration in the smaller sizes (i.e., for small j) and a very small particle concentration for particles of large diameter, as $\hat{S}_{N,j}$ increases steeply with increasing j. This does not affect the final distribution, but only takes a few more iterations for the iterative process to converge. Using a different starting solution based on the monodisperse sensitivities given by Liu and Pui (1979), no appreciable difference in the number of iterations for the procedure to converge were noticed.

For the iteration technique, a convergence criterion also must be specified. In general, a convergence criterion for an iterative procedure is a measure of the change of the retrieved function or solution between two consecutive iterations. For measurement instruments, the convergence criterion could also be a measure of how well the retrieved distribution fits the data. Several functions can be used as a measure of goodness of fit, among them being the sum of squares of relative differences (SSRD), as given by Equation (2.20):

$$\mathrm{SSRD} = \sum_{i=1}^{11} \left(\frac{\Delta I_i' - \Delta I_i}{\Delta I_i} \right)^2 \qquad (2.20)$$

where $\Delta I_i'$ is the calculated response for the ith channel based upon the test distribution and ΔI_i is the measured response, and the weighted least squares (WLS) as given by Equation (2.21):

$$\mathrm{WLS} = \sum_{i=1}^{11} \frac{1}{\sigma_i^2} \left(\frac{\Delta I_i'}{\sum_{k=1}^{11} \Delta I_k'} - \frac{\Delta I_i}{\sum_{k=1}^{11} \Delta I_k} \right)^2 \qquad (2.21)$$

where σ_i is the uncertainty in the measured ΔI_i. The σ_i depends on the instrumental uncertainty and the stability of the measured aerosol. As no a priori estimates of σ_i are available, it is assumed that the σ_i^2 is proportional to the normalized measured current difference. The proportionality constant is assumed to be unity for minimization purposes, as only the relative values of WLS are of interest.

In the present work, Equation (2.21) was used. Figure 2.10(a) shows the change in WLS with the number of iterations, using Equation (2.19)

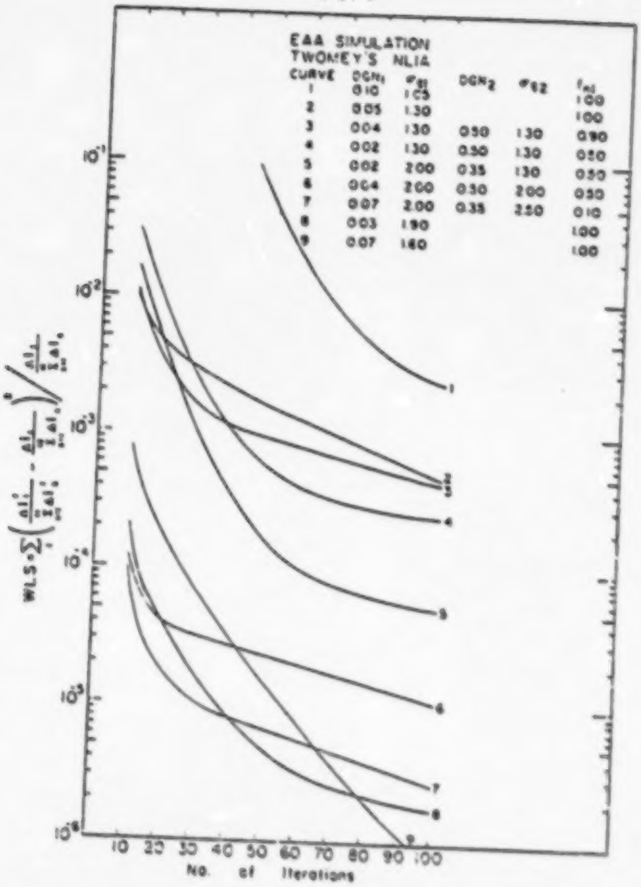

Figure 2.10(a) WLS vs. number of iterations for simulated EAA data, using Twomey's NLIA for data reduction.

as a starting guess. The response of the EAA for several log-normal distributions (both unimodal and bimodal log-normal) was first calculated using Equation (2.17), with $\varepsilon_i = 0.0$. These calculated current differences were then used as input to Twomey's NLIA. Using Equation (2.19) as the starting solution, the decrease in weighted least squares with the number of iterations was determined, and the results from some of the simulations are plotted on Figure 2.10(a). As can be seen from Figure 2.10(a), the WLS initially drops rapidly, but the change of WLS with the number of iterations decreases, as the number of iterations increase. Knutson (1977) found that after 20 iterations, the change in size distribution was negligible and so terminated the process after 20 iterations. In the present study, at first 20 iterations were carried out; then WLS according to Equation (2.21) was computed. If the value of WLS was less than 0.005, the iterative process was terminated. Otherwise, more iterations in increments of 5 were done, until the desired value of WLS was reached. After a maximum of 100 iterations, the procedure was halted even though WLS was not below the set value of 0.005. From simulation studies, it was found that 20 iterations were sufficient for most of the aerosol size distributions, with only a small fraction (about 5%) needing more than 20 iterations for reaching the convergence criterion. When the iterative procedure has converged, a set of 45 values of ΔN_j are determined, which fit the data acceptably. Equations (2.22) and (2.23) are then used to determine the surface and volume distribution of the aerosol.

$$\Delta S_j = \pi (D_{pj})^2 \, \Delta N_j \qquad (2.22)$$

$$\Delta V_j = \frac{\pi}{6} (D_{pj})^3 \Delta N_j \qquad (2.23)$$

where D_{pj} is the diameter of the particle in the jth size division. It should be noted here that the same results would have been obtained if ΔS_j or ΔV_j were used in Equation (2.18) instead of ΔN_j. In order to use ΔS_j, $\dot{S}_{S,j}$ would have to be used instead of $\dot{S}_{N,j}$ in Equation (2.18), and similarly, $S_{V,j}$ must be used for $S_{N,j}$.

Similarly, the NLIA(II) when applied to Equation (2.17) becomes:

$$\Delta N_j^{\,p} = \Delta N_j^{\,p-1} \times \sum_{i=1}^{11} R_{ij}\, \gamma_i^{\,p-1} \qquad j=1,\ldots 45 \qquad (2.24)$$

The starting solution used for NLIA(II) is given by Equation (2.19). Equation (2.21) is used as a measure of goodness of fit. Figure 2.10(b) shows the change of WLS with the number of iterations using Equation (2.19) as the starting solution. It can be seen from Figure 2.10(b) that the WLS vs. the number of iteration curves for different test distributions are very similar to Twomey's NLIA.

2.6.2.2.1 Simulation Studies for NLIA's

To test the NLIA's and determine their utility for solving the EAA data reduction problem, a simulation study was carried out. This study consisted of two parts. In the first part, the response of the EAA for several (approximately 250) log-normal distributions (both unimodal and bimodal) was first calculated. Table 2.3 gives the values of the bimodal log-normal distribution parameters which were used in all possible combinations for simulation. These calculated responses were then used as input for the NLIA's, and the solution determined.

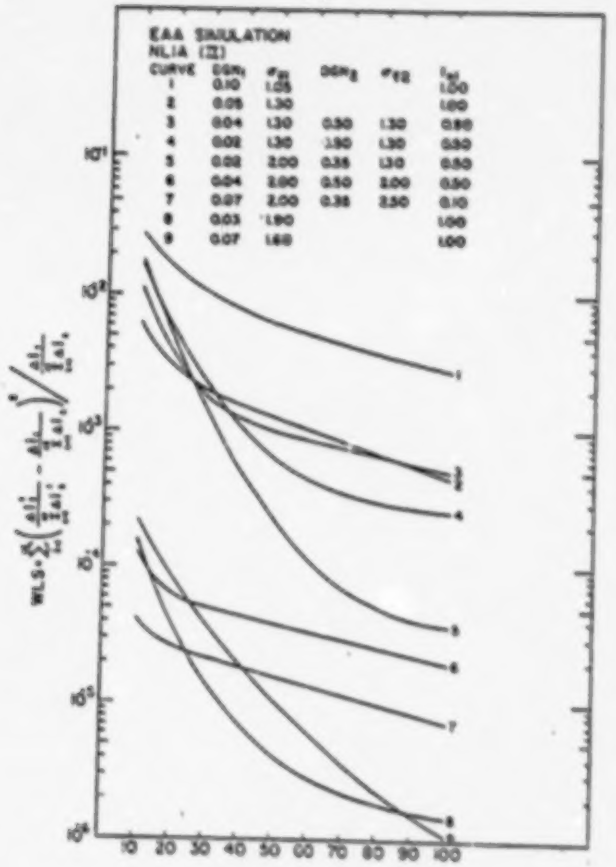

Figure 2.10(b) WLS vs. number of iterations for simulated EAA data, using NLIA(II) for data reduction.

Table 2.3 Values of the bimodal log-normal distribution parameters used in the simulation studies for the EAA

DGN_1 μm	σ_1	DGN_2 μm	σ_2	f_{n_1}
0.02	1.3	0.2	1.3	0.1
0.04	2.0	0.35	2.0	0.5
0.07	2.5	0.5	2.5	0.9

In addition to the bimodal aerosols, simulation studies were also carried out for unimodal and monodisperse aerosols of geometric mean diameters corresponding to the EAA channel midpoints. Figures 2.11, 2.12, and 2.13 show the results of two simulation runs.

It can be seen that for all three weightings, i.e., number, surface, and volume, the detailed distributions exhibit a highly oscillatory nature, which is not true of the input distribution. This was found for all the simulation runs made for the EAA using NLIA's. Also, NLIA's as applied to the EAA give 45 values of ΔN_j, even though only 11 measurements were made by the EAA. Thus, the retrieved distribution, in effect, has more "information" than was actually used as input to the data reduction procedure. In order to avoid both these inconsistencies, the ΔN_j's are then averaged over an interval of diameter, such that a histogram of 10 bars is obtained. The bounds for the histogram were chosen so as to coincide with the channel boundaries of the EAA as given by Liu and Pui (1975). It can be seen from Figures 2.11(a) and (b) that the retrieved histogram is very close to the input distribution. For Figures 2.12 and 2.13 [(a) and (b)], there is a discrepancy between the histogram and the input distribution in the last channel. This was observed for all the simulation runs done with the EAA for this method of data reduction. Similar results were obtained in several simulation runs done with ε_i as a random normal deviate with mean 0.0 and standard deviation of 5% of ΔI_i. The computer program for EAA data reduction using Twomey's NLIA, with an explanation of appropriate variables, is given in Appendix B.

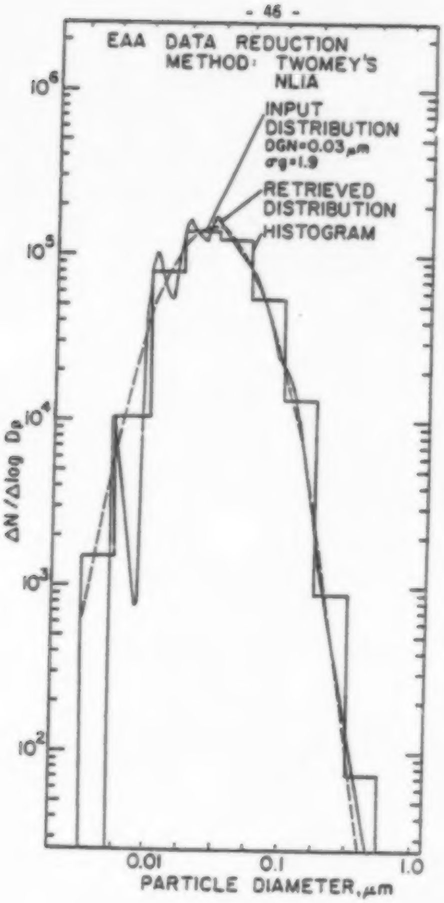

Figure 2.11(a) Retrieved number distribution using Twomey's NLIA for simulated response of the EAA to a unimodal log-normal distribution (DGN = 0.03 μm, σ_g = 1.9).

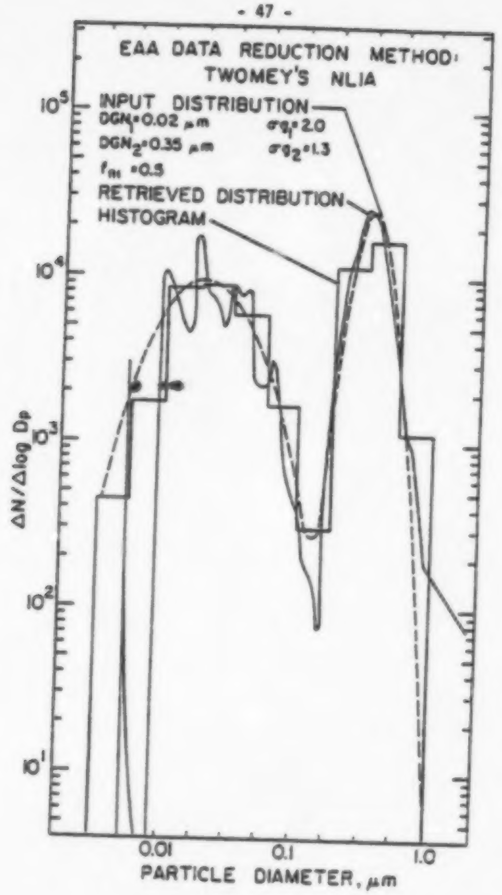

Figure 2.11(b) Retrieved number distribution using Twomey's NLIA for simulated response of the EAA to a bimodal log-normal distribution ($DGN_1 = 0.02$, $\sigma_{g1} = 2.0$, $DGN_2 = 0.35$, $\sigma_{g2} = 1.3$, $f_{n1} = 0.5$).

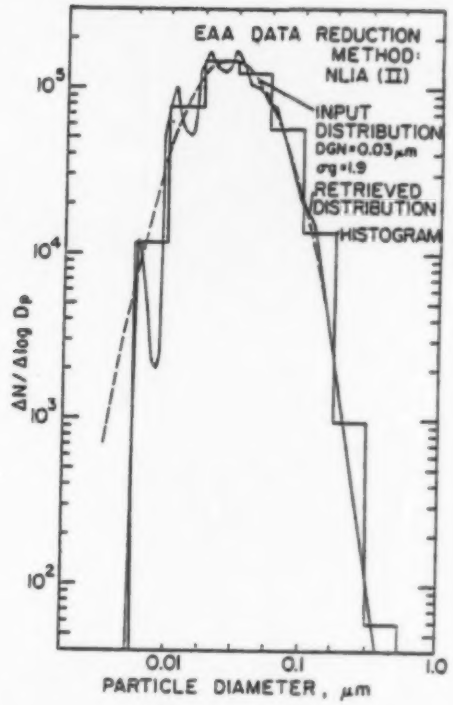

Figure 2.11(c) Retrieved number distribution using NLIA(II) for simulated response of the EAA to a unimodal log-normal distribution (DGN = 0.03 μm, σ_g = 1.9).

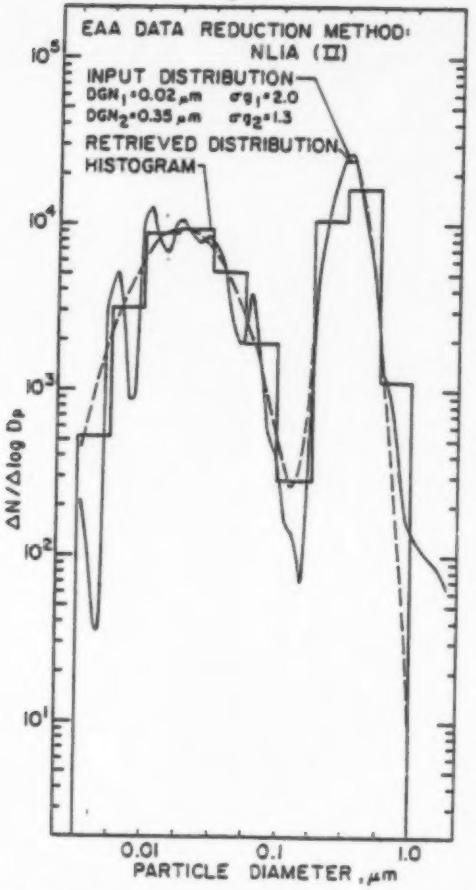

Figure 2.11(d) Retrieved number distribution using NLIA(II) for simulated response of the EAA to a bimodal log-normal distribution ($DGN_1 = 0.02$, $\sigma_{g1} = 2.0$, $DGN_2 = 0.35$, $\sigma_{g2} = 1.3$, $f_{n1} = 0.5$).

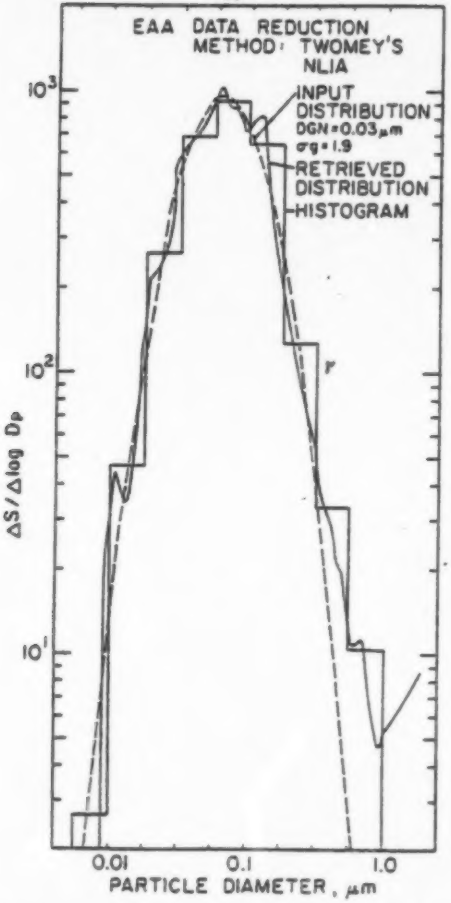

Figure 2.12(a) Retrieved surface distribution using Twomey's NLIA for simulated response of the EAA to a unimodal log-normal distribution (DGN = 0.03 µm, σ_g = 1.9).

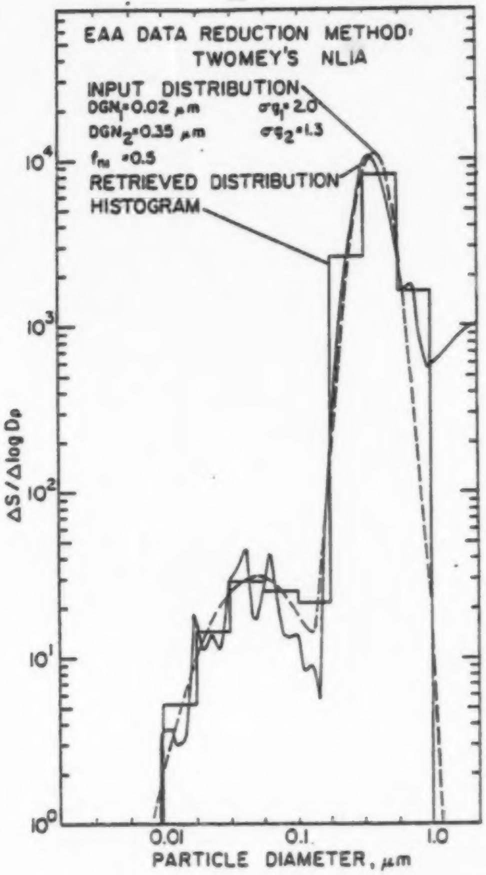

Figure 2.12(b) Retrieved surface distribution using Twomey's NLIA for simulated response of the EAA to a bimodal log-normal distribution (DGN_1 = 0.02, σ_{g1} = 2.0, DGN_2 = 0.35, σ_{g2} = 1.3, f_{n1} = 0.5).

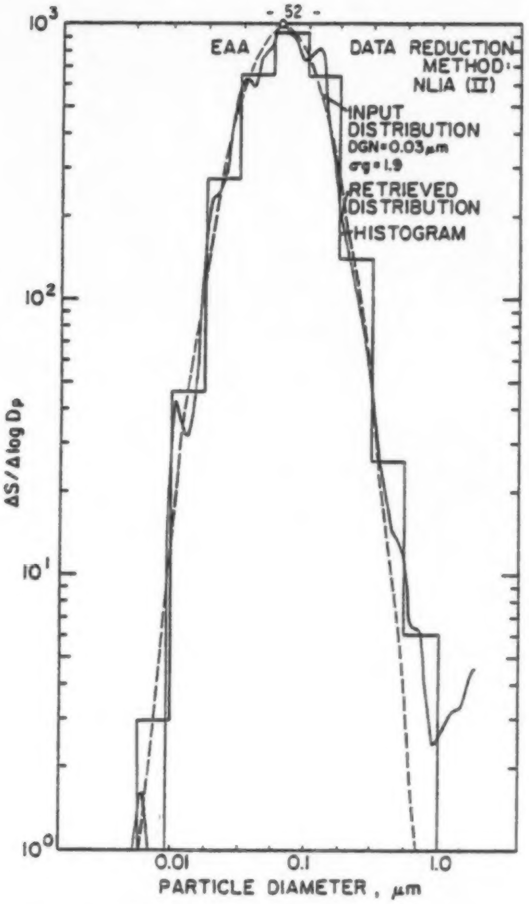

Figure 2.12(c) Retrieved surface distribution using NLIA(II) for simulated response of the EAA to a unimodal log-normal distribution (DGN = 0.03 μm, σ_g = 1.9).

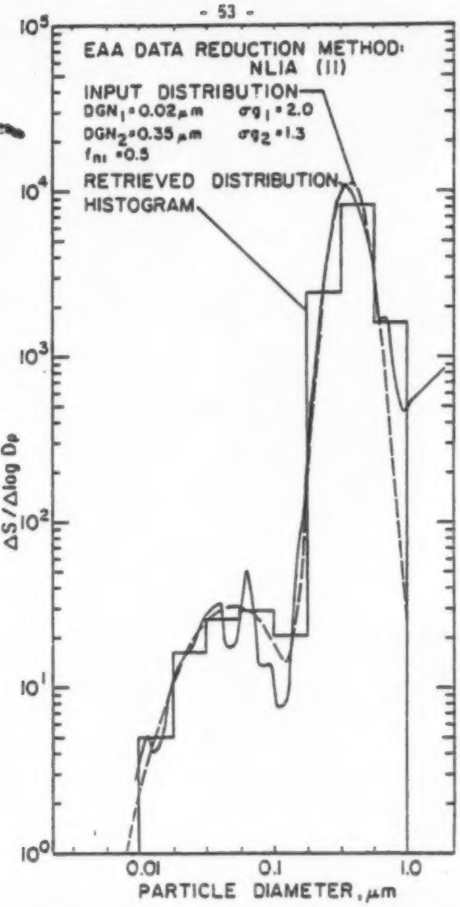

Figure 2.12(d) Retrieved surface distribution using NLIA(II) for simulated response of the EAA to a bimodal log-normal distribution (DGN_1 = 0.02, σ_{g_1} = 2.0, DGN_2 = 0.35, σ_{g_2} = 1.3, f_{n_1} = 0.5).

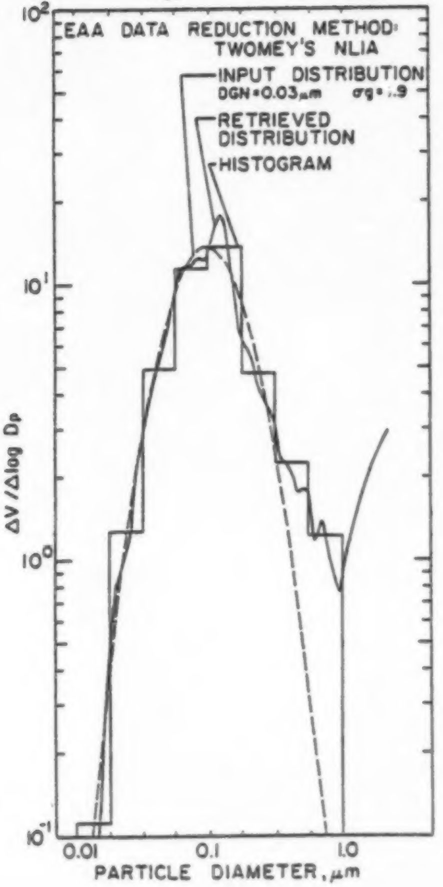

Figure 2.13(a) Retrieved volume distribution using Twomey's NLIA for simulated response of the EAA to a unimodal log-normal distribution (DGN = 0.03 µm, σ_g = 1.9).

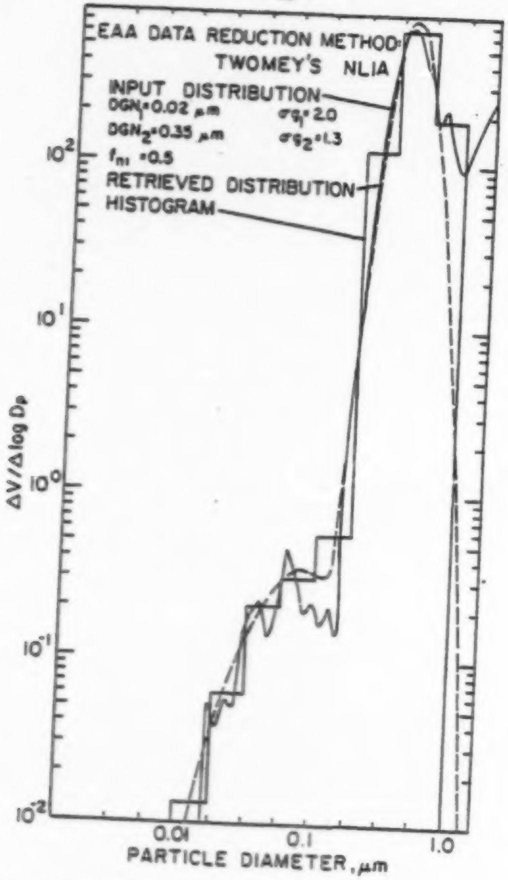

Figure 2.13(b) Retrieved volume distribution using Twomey's NLIA for simulated response of the EAA to a bimodal log-normal distribution (DGN_1 = 0.02, σ_{g1} = 2.0, DGN_2 = 0.35, σ_{g2} = 1.3, f_{n1} = 0.5).

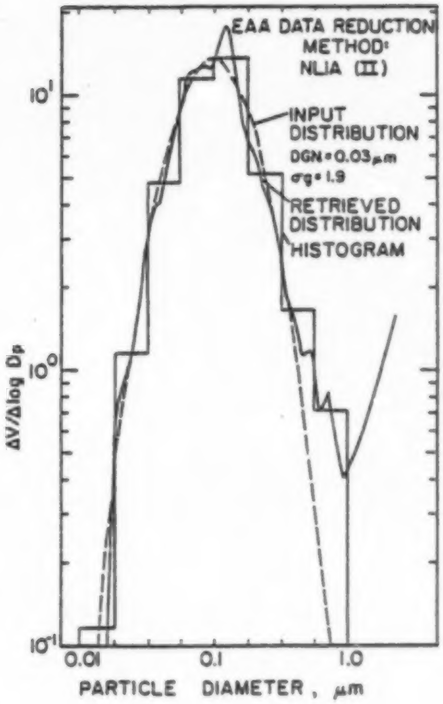

Figure 2.13(c) Retrieved volume distribution using NLIA(II) for simulated response of the EAA to a unimodal log-normal distribution (DGN = 0.03 μm, σ_g = 1.9).

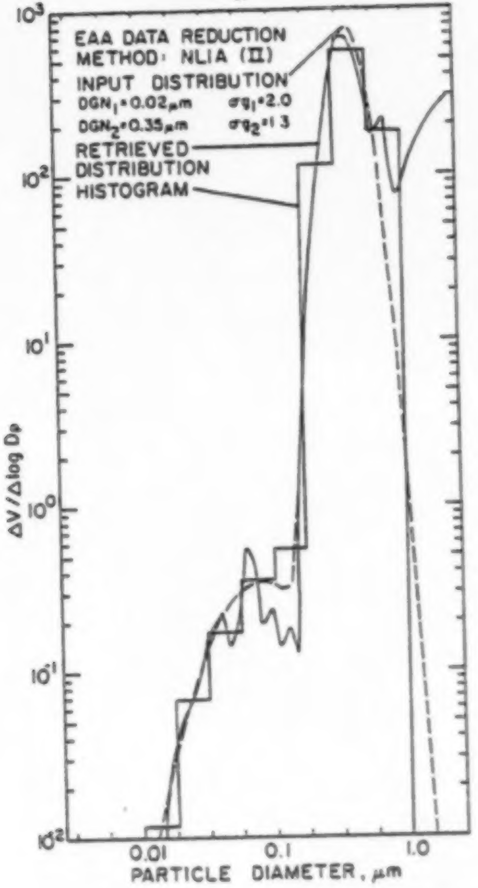

Figure 2.13(d) Retrieved volume distribution using NLIA(II) for simulated response of the EAA to a bimodal log-normal distribution (DGN_1 = 0.02, σ_{g1} = 2.0, DGN_2 = 0.35, σ_{g2} = 1.3, f_{n1} = 0.5).

Just as in Twomey's NLIA, for NLIA(II), the 45 values of ΔI_j are averaged over intervals of diameter. From Figures 2.11, 2.12, and 2.13 [(c) and (d)], it can he seen that the retrieved histogram is very close to the input distribution. This was observed for several simulation runs done for the EAA using NLIA(II). For both Twomey's NLIA and NLIA(II), the surface and volume concentration estimates for the last two channels were observed to be consistently higher than the input distribution for all the simulation runs. The reason for this lies in the fact that the electrical mobility vs. particle size curve has a small slope for large diameters. The computer program, with an explanation of appropriate variables, is given in Appendix B.

For the second part of the simulation study, a random normal error ϵ_i with mean 0.0 and a standard deviation of 5% of ΔI_i, $i=1,\ldots 11$ was added to the calculated ΔI_i's to simulate the instability of the measured aerosol and measurement error. These modified sets of ΔI_i's were then used as input for the NLIA's, and the retrieved distributions were compared with the results from the first part of the simulation study and were found to be within ±10% (corresponding to ±2   limits).

2.6.2.3 Simplex Minimization Method for Solving FIE of the First Kind

An approximate solution to FIE can also be obtained by fitting a function of a specified form to the observed data. After the fitting procedure is carried out, parameters which best fit the data are obtained. The function with the optimized parameters can then be taken as an approximate solution to the FIE or constants can he calculated based upon the optimized function, which are then used to obtain the final solution.

In order to use this method, the following three requirements have to be specified: (1) the form of the solution which can be specified by several parameters, (2) the minimization procedure which gives the set of parameters which best fit the data, and (3) the function to be minimized (hereafter referred to as the objective function).

Several mathematical functions have been described in the literature which model atmospheric size distributions. For example, Junge (1952, 1969) introduced the power law distribution given by Equation (2.25):

$$\frac{dN}{d\log D_p} = C' D_p^{-\nu} \qquad D_{p_L} \leq D_p \leq D_{p_u}$$

or (2.25)

$$n(D_p) = \frac{dN}{dD_p} = C'' D_p^{-(\nu+1)}$$

where C' and C" are constants and ν is the power law exponent. Junge proposed a value of 3 for ν, for D_p greater than 0.2 µm, with the power law being applicable up to D_p = 200.0 µm. The power law cannot be used to model submicron aerosols, and thus has not been used in the present work.

Another model proposed by Deirmendjian (1969), given by Equation (2.26), called the modified gamma distribution, attempts to describe the entire size distribution with a single function:

$$n(D_p) = a_o \frac{D_p}{2}^{\alpha_o} e^{-b_o \frac{D_p}{2}^{\gamma_o}}$$

and (2.26)

$$N_T = a_o \gamma^{-1} b_o^{-\frac{\alpha_o + 1}{\gamma_o}} \quad \frac{(\alpha + 1)}{\gamma_o}$$

where a_0, α_0, b_0, and γ_0 are the parameters of the distribution, and Γ is the gamma function.

Both the power law distribution and the modified gamma distribution do not describe adequately the size distribution of atmospheric aerosols, which is observed to have a multimodal aerosol size distribution, especially in surface and volume weightings. For atmospheric aerosols and many laboratory-generated aerosols, the log-normal distribution is better suited. The log-normal distribution given by Equation (2.27) for number distribution has a form which is independent of the size distribution weighting:

$$n(D_p) = \frac{N_T}{\sqrt{2\pi}\ \ln \sigma_g} \frac{1}{D_p} \exp^{-1/2} \quad \frac{\ln D_p/DGN}{\ln \sigma_g} \quad 2$$

or (2.27)

$$\frac{dn(D_p)}{d\ln D_p} = \frac{N_T}{\sqrt{2\pi}\ \ln \sigma_g} \exp^{-1/2} \quad \frac{\ln D_p/DGN}{\ln \sigma_g} \quad 2$$

where DGN is the number geometric diameter in μm and σ_g is the geometric standard deviation. The log-normal distribution being invariant under power transformation, the surface distribution is given by:

$$\frac{dS}{d\ln D_p} = \frac{S_T}{\sqrt{2\pi}\ \ln \sigma_g} \exp^{-1/2} \quad \frac{\ln D_p/DGS}{\ln \sigma_g} \quad 2 \qquad (2.28)$$

where DGS is the surface geometric mean diameter in μm, S_T is the total surface area concentration (μm^2/cc), and the volume distribution is given by

$$\frac{dV}{d\ln D_p} = \frac{V_T}{\sqrt{2\pi}\ \ln \sigma_g} \exp^{-1/2} \quad \frac{\ln D_p/DGV}{\ln \sigma_g} \quad 2 \qquad (2.29)$$

where DGV is the volume geometric mean diameter in μm, and V_T is the total volume concentration ($\mu m^3/cc$). The relationship between the number, surface, and volume geometric mean diameters is given by Equation (2.30):

$$\ln DGK = \ln DGN + K (\ln \sigma_g)^2 \tag{2.30}$$

where

if K = 0, DGK is the geometric number mean, DGN

if K = 2, DGK is the geometric surface mean diameter, DGS,

if K = 3, DGK is the geometric volume mean diameter, DGV,

and the total number, surface, and volume are related as follows:

$$C_K = \alpha_K N_T (DGN)^K \exp \left(\frac{K^2}{2} (\ln \sigma_g)^2\right) \tag{2.31}$$

where

for K = 0, C_K is N_T and α_K is 1,

for K = 2, C_K is S_T and α_K is π for spherical particles, and

for K = 3, C_K is V_T and α_K is $\pi/6$ for spherical particles.

Moreover, the representation of the aerosol size distribution by a log-normal function also has a physical basis. In aerosols, the size of particles is determined by processes whose effect on the size is proportional to the size itself, i.e., if there is a small particle, the process generally has a small effect on the particle size, while for a large particle, the effect is considerable. Such processes generally evolve log-normal distributions for particle size. In order to account for the multimodal nature of the distributions, Whitby (1975) proposed a composite distribution made of three log-normal distributions for representing atmospheric aerosols in the range 0.002 μm to about 60.0 μm. Whitby has since analyzed and successfully fitted data from field measurements done by the Particle Technology Laboratory of the University of Minnesota with trimodal log-normal distribution. For the present work, since both the EAA and the diffusion battery measure only submicron aerosols, a bimodal log-normal distribution was found to be sufficient.

The minimization procedure used in this thesis is based on the method developed by Nelder and Mead (1965). In this method, a function of m variables is minimized using a direct search method.

For a function of n variables, a general simplex of $n+1$ vertices, denoted by P_j, $j=1,2,\ldots m+1$ is initially formed and the objective function evaluated at each of the vertices. The value of the objective function at P_j is denoted by y_j. The vertex which has the highest value of the objective function (P_h) is then replaced by another point. To determine the new point, first the centroid ($\overline{P}$) of the remaining points (i.e., P_j, $j=1,2,\ldots m+1, j \neq h$) is calculated and the location of the next point is determined by one of the three operations: reflection, expansion, or contraction.

The coordinates of P^* determined by reflection are given by

$$P^* = (1 + \alpha)\,\overline{P} - \alpha\, P_h \tag{2.32}$$

where α is a positive constant. Thus, P^* lies on the far side of $\overline{P}$ from P_h, and on the line joining $\overline{P}$ and P_h. If y^* (value of the objective function at P^*) is between P_h and P_l (highest and lowest value of the objective function evaluated at the vertices of the simplex),

P_h is replaced by P^*, and a new simplex formed and the process restarted.

To show the process graphically, consider a two-variable function which is to be fitted. Let the two variables be x_1 and x_2. In Figure 2.14, P_1, P_2, and P_3 form the initial simplex, and assume that P_3 is P_h. The point $\bar{P}$ will then lie on the line joining P_1 and P_2, and P^* on the line joining $\bar{P}$ and P_3, and $P^*\bar{P}/P_h\bar{P} = \alpha$.

If P^* produces a new minimum, i.e., $y^* < y_1$, an expansion is attempted and P^{**} located. The coordinates of P^{**} are determined by

$$P^{**} = \quad P^* + (1 - \gamma)\,\bar{P} \tag{2.33}$$

and in Figure 2.14, $P^{**}\bar{P}/P^*\bar{P} = \gamma$, where γ is greater than unity. If y^{**} is less than y^*, P_h is replaced by P^{**}, a new simplex formed, and the process restarted. Otherwise, P_h is replaced by P^*.

On the other hand, a contraction is carried out, giving P^{***}, when reflection fails. The location of P^{***} is determined by

$$P^{***} = \beta\, P_h + (1 - \beta)\,\bar{P} \tag{2.34}$$

with the contraction coefficient β taking values between 0 and 1. In Figure 2.14, $P^{***}\bar{P}/P_h\bar{P} = \beta$. P^{***} replaces P_h, unless $y^{***} > \min\,(y_h, y^*)$, i.e., the contraction point is worse than the better of P_h and P^*. When contraction fails, all the P_j's are replaced by $P_j + P_1/2$, and the process restarted.

In this manner, the simplex continuously adapts itself to the landscape and contracts in the neighborhood of a minimum. The search process is halted when dimensions of the simplex become smaller than certain set values. In the present work, $\alpha = 1.0$, $\beta = 0.5$, and $\gamma = 2.0$ were chosen,

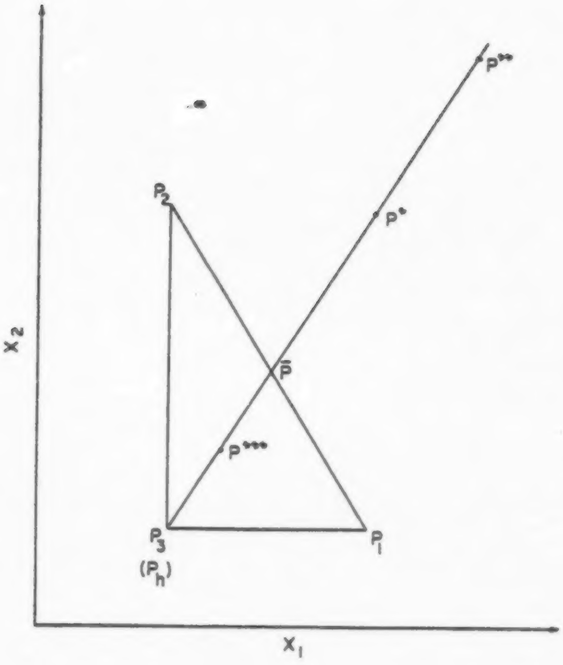
P^{**}
P^{*}
P_2
$\bar{P}$
P^{***}
P_3
(P_h)
P_1
X_2
X_1

Figure 2.14 Graphical representation of reflection, expansion, and contraction for two-dimensional space.

as recommended by Nelder and Mead (1965). It should be noted here that the minimum may not necessarily be a global minimum, but only a local minimum. Either the minimization process should be repeated with different starting simplices, or the solution can be taken as reasonably close to the global optimum if the value of the objective function is acceptable.

In this manner, the optimum value of the parameters which best fit the data are determined. The optimum test function can be taken as an approximate solution to the FIE, or constants based on the optimum parameters are determined and used with the measured data to calculate the final values of the solution.

The third requirement which must be specified if the simplex minimization method is to be used for data reduction is the form of the objective function. Various forms of the objective function are available which would measure the goodness of fit of the final solution with the data. Equations (2.20) and (2.21) describe two such objective functions which can be used. In the present work, Equation (2.21) has been used, as it takes into account the errors in the measured values of ΔI_i.

2.6.2.4 Simplex Minimization Method Applied to the EAA Data Reduction Problem

For the EAA data reduction problem, the simplex minimization method has been used extensively at the University of Minnesota and by other researchers in aerosol science.

For the EAA, the normalized bimodal log-normal distribution given in Equation (2.35) was used to model the solution:

$$f_N = \frac{dn\ (D_p)}{d\ln D_p} = f_{n_1} \frac{1}{\sqrt{2\pi}\ \ln \sigma_{g_1}} \exp^{-1/2} \frac{\ln D_p/DGN_1}{\ln \sigma_{g_1}}^{2}$$

$$+ \frac{(1 - f_{n_1})}{\sqrt{2\pi}\ \ln \sigma_{g_2}} \exp^{-1/2} \frac{\ln D_p/DGN_2}{\ln \sigma_{g_2}}^{2} \qquad (2.35)$$

where f_{n_1} is the number fraction in the first mode, DGN_1 and DGN_2 are the geometric number mean diameter in µm, and σ_{g1} and σ_{g2} are the geometric standard deviations for the two modes. For a bimodal log-normal distribution, $0 \leq f_{n_1} \leq 1$, while for a unimodal log-normal distribution, $f_{n_1} = 1.0$, with DGN_2 and σ_{g_2} being dummy parameters.

For the objective function, Equation (2.21) is used. The ΔI_i's are calculated using Equation (2.17) with $\varepsilon_i = 0.0$ and ΔN_j determined from Equation (2.35).

Using the simplex minimization algorithm, the optimum set of parameters which best fit the measured data are determined. In order to start the minimization procedure, an initial simplex must be formed. This can be accomplished by using as input a set of initial estimates for the parameters of the log-normal distribution.

The final value of the objective function can be used as a flag for determining whether the global minimum for the particular data set has been achieved. The smaller the value of the objective function, the better is the fit. We have found that if the final value of WLS is less than 0.1, the optimum set of parameters indeed represent the global minimum or are very close to it. Generally, a value of 0.05 can be obtained for the final value of WLS.

It should be noted here that the cost of computation can be made significantly less by choosing the initial estimates as close to the global minimum as possible, and that it is less expensive to reduce unimodal data compared to bimodal.

A criticism often leveled against using a curve fitting procedure, such as the simplex minimization method, for reducing the data of instruments measuring aerosol size distributions is that one has to make an assumption regarding the form of the size distribution. However, if the form of the function is general enough, such that it adequately represents the size distribution in the range of the instrument, this may not be a severe shortcoming. The bimodal log-normal function has five parameters, with up to two peaks and four inflection points. Thus, for the bimodal log-normal distribution, though the mathematical form of the function is fixed, its shape can be made to vary significantly by choosing different numerical values for its parameters. Moreover, as explained earlier, it is invariant under power transformations, and represents satisfactorily all three weightings, i.e., number, surface, and volume, in the size range of the EAA.

Once the optimum parameters have been determined, polydisperse sensitivities corresponding to the log-normal function with the optimum parameters are then calculated.

$$S_{N,i} = \frac{\Delta I_i'(*)}{\int_{\ln BD_i}^{\ln BD_{i+1}} f_N(DGN_1^*, \sigma g_1^*, DGN_2^*, \sigma g_2^*, f_{n_1}^*)\, d \ln D_p} \qquad (2.36)$$

$$\bar{S}_{S,i} = \frac{\Delta I_i'(*)}{\int_{\ln BD_i}^{\ln BD_{i+1}} f_S(DGS_1^*, \sigma_{g_1}^*, DGS_2^*, \sigma_{g_2}^*, f_{s_1}^*) \, d \ln D_p} \tag{2.37}$$

$$\bar{S}_{V,i} = \frac{\Delta I_i'(*)}{\int_{\ln BD_i}^{\ln BD_{i+1}} f_V(DGV_1^*, \sigma_{g_1}^*, DGV_2^*, \sigma_{g_2}^*, f_{v_1}^*) \, d \ln D_p} \tag{2.38}$$

where $\Delta I_i(*)$ is the calculated current for the optimized distribution in the ith channel, BD_i is the lower bound of the ith EAA channel, and BD_{i+1} is the upper bound of the ith EAA channel, both in microns. DGS_1^*, DGS_2^*, DGV_1^*, and DGV_2^* are determined using Equation (2.30), and $f_{s_1}^*$ and $f_{v_1}^*$ are determined using Equations (2.39) and (2.40), which are bimodal equivalents for the normalized form of Equation (2.31):

$$f_{s_1}^* = \frac{f_{n_1}^* (DGN_1^*)^2 \exp(2 (\ln \sigma_{g_1}^*)^2)}{f_{n_1}^* (DGN_1^*)^2 \exp(2 (\ln \sigma_{g_1}^*)^2) + (1 - f_{n_1}^*) (DGN_2^*)^2 \exp(2 (\ln \sigma_{g_2}^*)^2)} \tag{2.39}$$

and

$$f_{v_1}^* = \frac{f_{n_1}^* (DGN_1^*)^3 \exp(\frac{9}{2} (\ln \sigma_{g_1}^*)^2)}{f_{n_1}^* (DGN_1^*)^3 \exp(\frac{9}{2} (\ln \sigma_{g_1}^*)^2) + (1 - f_{n_1}^*) (DGN_2^*)^3 \exp(\frac{9}{2} (\ln \sigma_{g_2}^*)^2)} \tag{2.40}$$

With Equations (2.41), (2.42), and (2.43), the final size distribution is obtained.

$$\Delta N_i^* = \Delta I_i/\tilde{S}_{N,i} \qquad i=1,\ldots 10 \qquad (2.41)$$

$$\Delta S_i^* = \Delta I_i/\tilde{S}_{S,i} \qquad i=1,\ldots 10 \qquad (2.42)$$

$$\Delta V_i^* = \Delta I_i/\tilde{S}_{V,i} \qquad i=1,\ldots 10 \qquad (2.43)$$

The standard error of the parameters of the optimum log-normal distribution are proportional to the square root of the diagonal elements of the inverse of the Hessian (H) matrix (Nelder and Mead, 1965). The elements of the Hessian, h_{ij}, are given by Equation (2.44):

$$h_{ij} = \frac{1}{2}\frac{\partial^2 WLS}{\partial x_i\ \partial x_j} \qquad \begin{matrix} i=1,\ldots 5 \\ j=1,\ldots 5 \end{matrix} \qquad (2.44)$$

where x_i and x_j are the parameters of the fitted function, and WLS is the objective function.

The proportionality constant for σ_i^2 [Equation (2.21)] was assumed to be unity for the minimization procedure, as only the relative values of the objective function were of interest. For estimating the standard errors of the parameters, however, the absolute values of the second derivative must be determined. Thus, the proportionality constant for σ_i^2 must also be determined. The maximum σ_i^2, for all the EAA channels, for laboratory-generated aerosols was experimentally determined to be approximately 15% of the normalized measured current difference. Thus, the standard error estimate of the parameters of the optimal log-normal aerosol size distribution is 0.15 times the square root of the diagonal element of the inverse of the Hessian matrix, with the Hessian being evaluated under the assumption that the value of the proportionality constant is unity. This procedure gives reasonable estimates of standard error for the parameters of unimodal log-normal distributions; however, for the bimodal distribution, curvature of WLS hypersurface is small near the optimum point. Thus, the standard error estimates cannot be determined due to round-off errors in evaluating the Hessian.

2.6.2.4.1 Simulation Studies for Simplex Minimization Method

The simulation studies to test the applicability of the simplex minimization method to the EAA data reduction problem were also carried out in two parts. In the first part, the effect of initial estimates which are used for starting the search procedure was evaluated, while in the second part, the effect of random noise added to the calculated response of the EAA on the values of the fitted parameters was evaluated.

To investigate the effect of initial estimates on the method, two strategies were evaluated. For both strategies, the parameters for the input bimodal log-normal distribution were the various combinations of values of the five parameters given in Table 2.3. For the first strategy, a blind initial estimate ($DGN_1 = 0.0173$ μm, $\sigma_{g1} = 1.75$, $DGN_2 = .173$ μm, $\sigma_{g2} = 1.75$, and $f_{n1} = 0.5$) was used. With this initial estimate, about 60% of the distributions were discovered. For the remaining 40% of the cases, only local minima were obtained and the values of the parameters corresponding to the local minima were different from the values of the parameters for the input bimodal log-normal distribution. The final value of WLS for all the unsuccessful cases was always greater than 1×10^{-3}, and thus, it proved to be a useful flag to determine whether or not global minimum had been reached. For the second strategy, first an approximate size distribution was determined using the monodisperse sensitivities of Liu and Pui (1975). The size distribution was then divided into two regions, the first region corresponding to the first five channels of the EAA and the second region corresponding to the last five channels of the EAA. For each region, a geometric mean and a geometric standard deviation were calculated, assuming the size distribution to be a unimodal log-normal one. From the total concentration estimates in each region, the number fraction in the first mode was also estimated. These values of the parameters were used as initial estimates for the simplex minimization curve fitting method. For the second strategy, no significant improvement in the percentage of distribution recovered was noticed. Thus, for analyzing experimental data, initial estimates were entered as input in the computer program.

During this part of the simulation study, it was observed that if the two modes of the bimodal log-normal distribution are not distinctly separate, the same bimodal log-normal distribution could be represented very closely by more than one set of values for the five parameters. Figure 2.15 shows one example of such a distribution. Similar observations have also been reported by Fissan (1978).

For the second part of the simulation study for this method, normal random error of 5%, 10%, and 15% ΔI_i, $i=1,\ldots 11$ was added to the calculated response of several unimodal and bimodal log-normal distributions. These responses were then used as input to the simplex minimization method for EAA data reduction, and the retrieved parameters were determined. Table 2.4 gives the comparison between the parameters

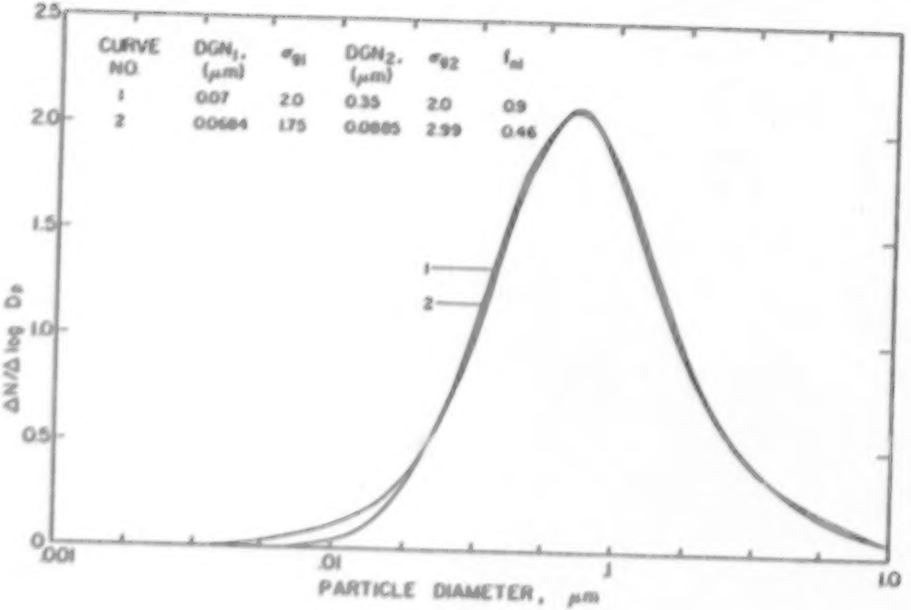

Figure 2.15 Overlapping bimodal log-normal distributions

Table 2.4 Comparison of the input and corresponding retrieved distribution parameters of log-normal aerosol distributions for simplex minimization method simulation studies for the EAA

Case	DGN_1	σ_{g_1}	DGN_2	σ_{g_2}	f_{n_1}	WLS
A1)	0.0300	1.90	---	---	1.0	---
B2)	0.0300	1.90	---	---	1.0	1.521×10^{-3}
C3)	0.0292	1.92	---	---	1.0	5.458×10^{-3}
D4)	0.0276	1.96	---	---	1.0	1.334×10^{-2}
A	0.0720	1.60	---	---	1.0	---
B	0.0707	1.60	---	---	1.0	5.229×10^{-3}
C	0.0696	1.58	---	---	1.0	1.922×10^{-2}
D	0.0692	1.55	---	---	1.0	4.532×10^{-2}
A	0.1000	1.05	---	---	1.0	---
B	0.0980	1.04	---	---	1.0	1.799×10^{-4}
C	0.0980	1.04	---	---	1.0	1.799×10^{-4}
D	0.0969	1.04	---	---	1.0	3.772×10^{-3}
A	0.0450	1.30	---	---	1.0	---
B	0.0467	1.25	---	---	1.0	1.606×10^{-3}
C	0.0476	1.25	---	---	1.0	6.162×10^{-3}
D	0.0491	1.22	---	---	1.0	1.588×10^{-2}

(continued)

1)A: Input distribution

2)B: Retrieved distribution for 5% error case

3)C: Retrieved distribution for 10% error case

4)D: Retrieved distribution for 15% error case

Table 2.4 (continued)

Case	DGN_1	σ_{g1}	DGN_2	σ_{g2}	f_{n1}	WLS
A	0.0200	1.30	0.5000	1.30	0.50	---
B	0.0200	1.30	0.5000	1.30	0.50	3.756×10^{-4}
C	0.0192	1.34	0.5000	1.30	0.50	1.466×10^{-3}
D	0.0200	1.30	0.5000	1.30	0.50	3.334×10^{-3}
A	0.0200	2.00	0.3500	1.30	0.50	---
B	0.0177	2.21	0.3500	1.30	0.50	1.112×10^{-3}
C	0.0178	2.15	0.3554	1.22	0.47	1.084×10^{-3}
D	0.0144	2.22	0.3500	1.30	0.50	8.56×10^{-3}
A	0.0400	1.30	0.5000	1.30	0.90	---
B	0.0408	1.28	0.5000	1.30	0.90	1.275×10^{-3}
C	0.0424	1.26	0.4993	1.18	0.89	2.483×10^{-3}
D	0.0429	1.23	0.5000	1.30	0.90	1.115×10^{-2}
A	0.0400	2.00	0.5000	2.00	0.50	---
B	0.0392	2.17	0.5000	2.00	0.50	7.274×10^{-4}
C	0.0388	2.16	0.4828	1.85	0.49	9.232×10^{-4}
D	0.0389	2.56	0.5000	2.00	0.50	8.529×10^{-3}
A	0.0700	2.00	0.3500	2.50	0.10	---
B	0.0606	1.59	0.3500	2.50	0.10	4.359×10^{-4}
C	0.0680	1.99	0.3478	2.73	0.07	1.154×10^{-3}
D	0.0457	1.64	0.3650	2.58	0.10	5.710×10^{-3}

of the input and retrieved distribution with the corresponding WLS. As can be seen from the table, the simplex minimization method retrieves the input parameters satisfactorily, with the average discrepancy between the input and retrieved parameters being about 5% for the case with 15% error. As expected, the WLS increases with increase in % error for any single distribution. It should also be noted here that the value of WLS obtained for the 15% error case is of the same order of magnitude as the final value of WLS observed for experimental and field EAA data reduced by the simplex minimization curve-fitting method.

CHAPTER 3

DIFFUSION BATTERY: PRINCIPLE AND DATA REDUCTION

3.1 Introduction

This chapter is concerned with the diffusion battery method of aerosol size distribution measurement. A brief historical review is first given. This is followed by an explanation of the operating principle of the diffusion battery and the treatment of the data reduction problem for the device.

Townsend (1899) determined the diffusion coefficient of gaseous ions by passing a laminarly flowing ionized gas through a narrow tube and measuring the penetration of the ions. Since then, Nolan and Guerrini (1935) and Nolan and Nolan (1938) have applied the method to estimate the diffusion coefficient of atmospheric nuclei. They passed diluted atmospheric air, in a steady laminar stream, through a box which consisted of several rectangular channels, and determined the penetration of the nuclei using the Aitken Pocket Dust Counter. Knowing the penetration of the nuclei, the dimensions of the rectangular channels, and the flow rate, they determined the diffusion coefficient of the condensation nuclei. Using different flow rates and different dimensions for the rectangular channels, they found that the diffusion coefficient of the nuclei varied significantly. Pollak and Metnieks (1957) explained these variations in the diffusion coefficient by considering polydisperse aerosols. They showed theoretically that the equivalent diffusion coefficient of a polydisperse aerosol increases as the flow rate through the diffusion cell is increased. Thus, the "equivalent diffusion size" from a single measurement has little practical significance.

As the diffusion coefficient decreases rapidly with particle size, attempts have been made to determine the aerosol size distribution by making penetration measurements of laminarly flowing aerosols through several diffusion cells. Using one of the several data reduction procedures available, the set of penetration measurements are converted to the aerosol size distribution. To make these measurements, a battery of diffusion cells is required. Initially, these batteries consisted of either several clusters of tubes of different lengths and diameters or a set of rectangular channels with different geometric dimensions. These diffusion batteries were expensive and unwieldy to use, and thus have not been used extensively to measure the size distribution of submicron aerosols. Recently, Sinclair (1972) developed a portable diffusion battery about 2 feet in length, using "collimated hole structures" (CHS) (Brunswick Corp., 69 Washington St., Chicago, IL 60602) of different thicknesses for each cell. This innovation greatly improved the prospects of measuring the aerosol size distribution by the diffusion deposition method. The mechanical details of the battery which was used in the present study are given later in this chapter. Since then, Sinclair and Hoopes (1975) introduced a novel form of the diffusion battery, which consisted of a series of 635 mesh stainless steel screens instead of the cluster of tubes or rectangular channels. In this device, 55 4-cm. diameter screens are arranged in a series of groups, through which aerosol is drawn either at 4 or 6 lpm, and the penetration of submicron aerosol is measured with a condensation nuclei counter. Knowing the measured penetration and the calibration of the screen diffusion battery, the aerosol size distribution is determined using a data reduction procedure. The screen diffusion battery is commercially available from TSI, Inc. (P. O. Box 43394, St. Paul, MN 55164).

3.2 Diffusion Batteries

3.2.1 CHS Battery

The "collimated hole structure" (CHS) battery used in this study is based on the one developed by Sinclair (1972). The specific device used here consists of 18 stainless steel discs of 1.435 in. diameter and varying thickness. Each disc has approximately 9750 holes, with the nominal diameter of each hole being about 240 μm. The thickness of CHS varies from 1/8 in. to 1½ inches. Each disc is mounted into a sleeve with O-rings. The sleeves, together with an inlet and outlet funnel, are bolted together. The 18 CHS mounted in series are then divided into 11 groups, each group corresponding to a stage of the diffusion battery. Between each of the stages, a side tube is attached to measure the particle concentration with a CNC. As it is impossible to mount the CHS such that the holes coincide, each disc is separated by a space of ¼ in., with a ½ in. spacing between two discs that have a side tube soldered between them for measuring particle concentrations. Table 3.1 gives the thickness of the CHS in each stage of the diffusion battery. Figure 3.1 is a photograph of the collimated hole structure (magnification = 40), and Figure 3.2 shows a schematic of the CHS battery.

Table 3.1 CHS thickness in diffusion battery stages

Stage No.	Nominal CHS thickness in the stage (inches)
1	1/8
2	1/8
3	1/4
4	3/8
5	1/2
6	3/4
7	1.0,
8	1.0, 1/2
9	1.0, 1.0
10	1.0, 1.0, 1.0
11	1.0, 1.0, 1.0, 1.0

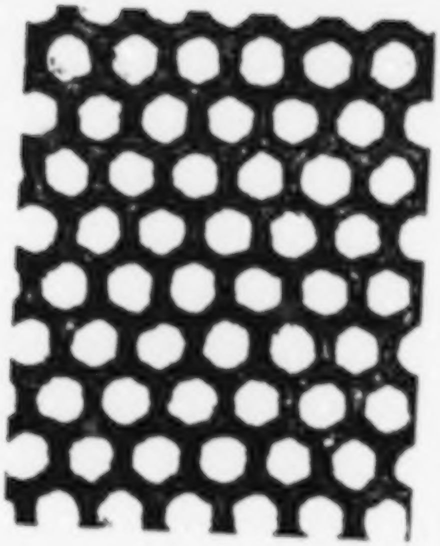

Figure 3.1 Collimated hole structure (CHS) (magnification = 40).

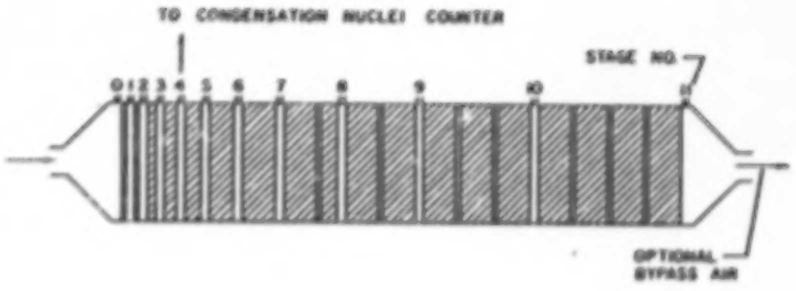
TO CONDENSATION NUCLEI COUNTER
STAGE NO.
0
1
2
3
4
5
6
7
8
9
10
11
OPTIONAL BYPASS AIR

Figure 3.2 Schematic diagram of CNS diffusion battery.

3.2.2 TSI Screen Diffusion Battery

The design of the TSI screen diffusion battery is based on the one developed by Sinclair and Hoopes (1975). It consists of 55 4-cm. diameter 635 mesh stainless steel screens placed in series in 10 groups or stages. The first stage consists of one screen; the second stage, of two; and so on up to the tenth stage, which consists of ten screens. Figure 3.3 (courtesy TSI, Inc.) gives a schematic of the screen battery. The input concentration is measured from port 0, and by making concentration measurements through each of the remaining 10 ports, the penetration or the diffusional loss in each of the stages can be determined.

3.3 Condensation Nuclei Counters

To measure the penetration of the aerosol through the various stages of the diffusion battery, an aerosol sensor is needed. Generally, for submicron aerosols, condensation nuclei counters (CNC) are used to measure the total particle concentration at the exit of the various stages of the diffusion battery. Aitken in 1890-1891 introduced the "Aitken Pocket Nuclei Counter" to measure the total concentration of submicron aerosols. Since then, various other devices have been developed and tested for measuring the concentration of submicron aerosols. One of these is a counter originally developed by Nolan and Pollak (1946), and further developed by Pollak and Metnieks (1957). This counter is referred to as the Pollak counter in the literature, and until recently was used as the standard for calibrating other condensation nuclei counters. Several nuclei counters are also available commercially. A simple manually-operated device is available from Gardner

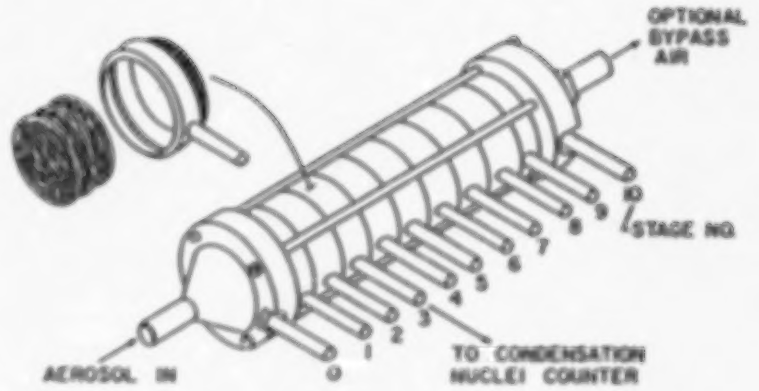
OPTIONAL BYPASS AIR
STAGE NO.
10
9
8
7
6
5
4
3
2
1
0
TO CONDENSATION NUCLEI COUNTER
AEROSOL IN

Figure 3.3 Schematic diagram of TSI screen diffusion battery (courtesy TSI, Inc.).

Associates (3436 Carman Road, Schenectady, NY 12303), and the Pollak counter, from GSI, Inc. (1254 Main Street, Waltham, MA 02154). Fully automatic nuclei counters are available from General Electric Company (Electronic Systems Division, Ordnance Systems, 100 Plastics Ave., Pittsfield, MA 01201), and also from Environment/One Corp. (Balltown Road, Schenectady, NY 12309).

In the CNC, the aerosol is saturated with water vapor and then expanded adiabatically to a lower pressure, producing supersaturation. In the supersaturated state, the water vapor condenses on the particles, forming droplets. These are then measured optically by light scattering or by light transmission principles.

The CNC is used to measure the total concentration of particles in the submicron range. The CNC was believed, until recently, to be capable of detecting particles as small as 25 Å over a wide range of concentrations from 100 particles/cc to 10^7 particles/cc. Recently, Liu and Kim (1977), Bricard et al. (1977), Cooper and Langer (1978), and Porstendorfer and Soderholm (1978) have found that the response of the automatic condensation nucleus counters is a function of particle size below 0.25 μm diameter. Thus, there is considerable ambiguity as to what is the smallest detectable size for the CNC.

The automatic counters described above have pulsating input flow. The diffusion deposition equations mentioned later in this chapter are valid for steady laminar flow, and thus, to use these CNC's, an anti-pulsator has to be used downstream of the diffusion battery, just before the CNC. Such an anti-pulsator was developed by Sinclair (1977) which reduced the fluctuations in the flow due to the CNC to a negligible value. Recently TSI, Inc. introduced commercially a continuous flow condensation nucleus counter. This CNC is based on two continuous flow nucleus counters, one developed by Bricard et al. (1976), and the other, by Sinclair and Hoopes (1975). The TSI CNC is described in detail by Agarwal et al. (1977).

Figure 3.4 gives the schematic diagram of the TSI continuous flow condensation nucleus counter used in this study. The aerosol whose concentration is to be measured first flows over a pool of N-butyl alcohol maintained at 35°C. The saturated aerosol then enters the condensation tube, which is kept at a temperature of 10°C by a thermoelectric cooling module. The alcohol vapor condenses on the particles in the condensor tube, forming droplets. Near the exit of the condenser tube, the size of the droplets is about 10 um. The droplets then enter the detector. For particle concentrations below 1000 particles/cc (single particle counting mode), each particle produces a pulse from the scattered light when it traverses the viewing volume. In this mode, the particle concentration is estimated from the number of pulses counted per unit time and the aerosol flow rate (5 cc/sec). Thus, no further calibration is required. However, corrections for coincidence error and statistical particle count accuracy must be made to the final readings. Agarwal et al. (1977) recommend the procedure described by Willeke and Liu (1976) for coincidence error and particle count accuracy in optical particle counters. The ratio of the indicated particle concentration, N_I, to the true

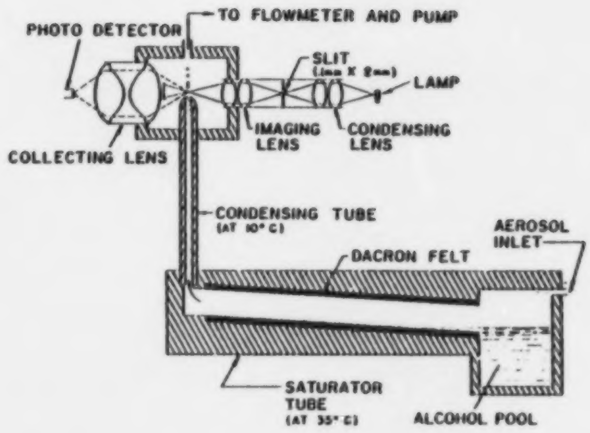
PHOTO DETECTOR
TO FLOWMETER AND PUMP
SLIT (1mm X 2mm)
LAMP
IMAGING LENS
CONDENSING LENS
COLLECTING LENS
CONDENSING TUBE (AT 10° C)
AEROSOL INLET
DACRON FELT
SATURATOR TUBE (AT 35° C)
ALCOHOL POOL

Figure 3.4 Schematic diagram of TSI continuous flow condensation nucleus counter (CNC) (courtesy TSI, Inc.).

particle concentration, N_0, is given by Equation (3.1) (from Willeke and Liu):

$$\frac{N_I}{N_o} = e^{-v'N_o} \qquad (3.1)$$

where v' is the viewing volume. The error due to particle count accuracy has been made to within 10% for a concentration of 0.1 particles/cc by considering longer averaging times (200 sec.) for very small particle concentrations.

For particle concentrations greater than 1000 particles/cc (photometric mode), the DC signal produced by the scattered light due to particles in the viewing volume is measured. The DC signal is converted to particle concentration by an electronic eight-segment linearizer, which incorporates the calibration characteristics of the CNC in the photometric mode.

The unit supplied to the University of Minnesota by TSI had a 20% discrepancy between the single particle and the photometric mode for a particle concentration around 1000 particles/cc. Thus, for the present study, only the single particle counting mode was used to measure the penetrations from the various stages of the diffusion batteries.

To verify the concentration measured by the TSI CNC in the single particle counting mode, comparisons between the particle concentrations measured by the CNC and the electrical classification method, described by Liu and Pui (1974), were made. Figure 3.5 gives the schematic of the CNC calibration set-up. Polydisperse NaCl aerosol was generated by a Collison atomizer. This aerosol was passed through a diffusion dryer,

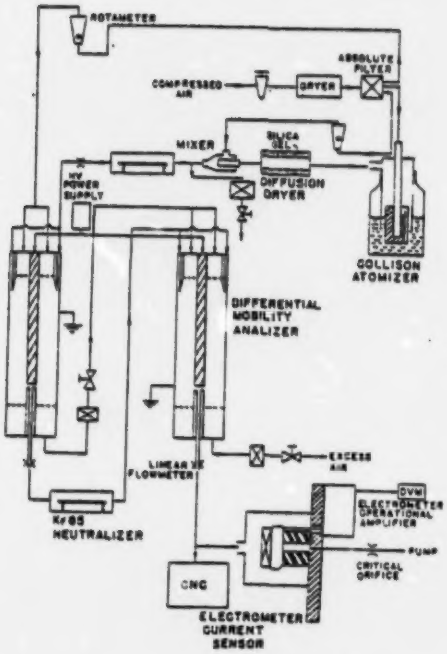
ROTAMETER
ABSOLUTE FILTER
COMPRESSED AIR
DRYER
SILICA GEL
MIXER
HV POWER SUPPLY
DIFFUSION DRYER
COLLISON ATOMIZER
DIFFERENTIAL MOBILITY ANALIZER
EXCESS AIR
LINEAR FLOWMETER
DVM
ELECTROMETER OPERATIONAL AMPLIFIER
Kr 85 NEUTRALIZER
PUMP
CRITICAL ORIFICE
CNC
ELECTROMETER CURRENT SENSOR

Figure 3.5   Schematic diagram of the TSI CNC calibration set-up.

a Kr-85 neutralizer, and it then entered a differential mobility analyzer (DMA). Positively, singly charged monodisperse particles exit from the DMA. To avoid making corrections for doubly charged particles, the relatively monodisperse output aerosol from the first DMA was used as an input to the second DMA. The output aerosol from the second DMA was highly monodisperse, with a negligible fraction of doubly charged particles. The monodisperse aerosol from the second DMA was simultaneously sampled by the CNC and an electrometer. The current measured by the electrometer is related to the input concentration N by Equation (3.2):

$$I = N\,e\,q \qquad (3.2)$$

where e is the elementary unit of charge ($e = 1.6 \times 10^{-19}$ coulombs) and q is the electrometer volumetric flow rate. Calibration checks were performed using NaCl particles of 0.056 μm, 0.042 μm, 0.036 μm, and 0.024 μm diameter. The particle concentration measured by the CNC was found to be within 5% of the concentration measured by the electrometer.

The lower and upper particle size limits for the TSI CNC have not yet been established, but it is assumed that the upper limit is around 1.0 μm, as particles greater than 1.0 μm may be lost in the plumbing of the instrument. For the lower size limit, the "counting efficiency" (term used by Liu and Kim, 1976) of the TSI CNC is nearly 100% down to 0.024 μm. Agarwal (1979) has verified that the counting efficiency is approximately 100% for 0.01 μm particles also. In the present study, the counting efficiency was assumed to be 100% for particles of diameter down to 0.0032 μm for calculation purposes. This does not alter the measurements and the final results significantly, as all the aerosols measured had a negligible concentration of particles below 0.01 μm in diameter. The calculation procedures developed in the present work are general, and can be applied to aerosols which have significant concentrations of particles smaller than 0.01 μm in diameter. For such cases, the appropriate counting efficiencies for the CNC must be taken into account. Though the CNC can measure particles up to 1.0 μm in diameter, the predicted penetration of 1.0 μm particles, taking into consideration only diffusion losses while neglecting settling due to gravity, is on the high side. Thus, to get more accurate estimates of the number concentration of larger particles, settling due to gravity must be considered. In the present work, the concentration of particles greater than 0.56 μm was negligible. Thus, the assumption of neglecting the losses due to gravity had no substantial effect on the measurements and results.

3.4 Operating Principle

As mentioned before, in the diffusion battery method, an aerosol is first passed through a series of diffusion cells in a laminar stream, and the penetration of aerosol for the different cells is measured. When the aerosol passes through a diffusion cell, due to the random Brownian motion, the particles collide with the walls and adhere to it. Small particles have a large diffusion coefficient and therefore have a small penetration as compared to large particles. Figure 3.6 shows the variation of the diffusion coefficient with the particle diameter. It can be seen from this curve that the diffusion coefficient changes by five orders of magnitude when the particle diameter changes by three decades.

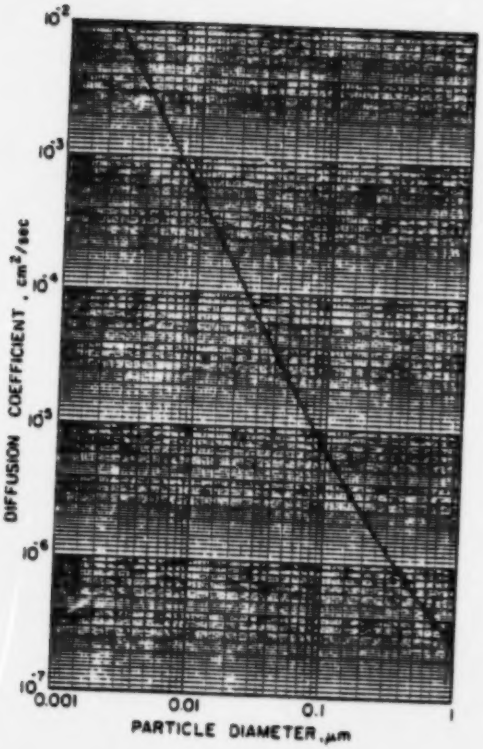

Figure 3.6 Diffusion coefficient vs. particle diameter.

Thus, in order to use the diffusion battery method, the penetration of the aerosol through a diffusion battery should either be predicted theoretically or estimated by a calibration matrix, which gives the penetration or the diffusion loss of monodisperse aerosols of various sizes through each diffusion cell.

For circular tubes or rectangular channels, theoretical penetrations have been determined by several investigators (Townsend, 1899; Gormley and Kennedy, 1949; Davies, 1946; Tan and Hsu, 1970, 1971; Smutek, 1971; and Ingham, 1975). The theoretical penetrations have been derived under the assumptions that the flow is laminar and Poisseulle, with no formation of aerosol in the flowing field. It is also assumed that the losses due to settling and mass transfer along the tube in the direction of the flow are negligible. The last assumption is valid for Peclet number, Pe, >> 1.0, where $Pe = Q/\pi dD$, with Q being the volumetric flow rate through the tube or channel; d, the characteristic dimension of the tube (diameter) or channel (half-width); and D, the diffusion coefficient of the particle.

Davies (1946) obtained Equation (3.3) for penetration of aerosols in tubes:

$$P(D_p) = 0.819 \exp(-14.63\Delta) + 0.0976 \exp(-89.22\Delta) + 0.019 \exp(-212\Delta) + \ldots \qquad (3.3)$$

where $P(D_p)$ is the penetration of particle size D_p, and $\Delta = \pi DL/4Q$, with L being the length of the tube and Q the volumetric flow rate through the tube. The diffusion coefficient D, for a particle of size D_p is given by Equation (3.4) (from Fuchs, 1966):

$$D = \frac{k\ T}{3\pi\ \mu\ D_p}\quad 1 + \frac{2A\ l}{D_p} + \frac{2B\ l}{D_p} \exp\ (-b\ D_p/2l) \qquad (3.4)$$

where $A = 1.246$, $B = 0.42$, and $b = 0.87$ are constants, l is the mean free path of the air molecules, k is the Roltzmann constant, T is the absolute temperature, and μ is the viscosity of air.

Gormley and Kennedy (1949) obtained the expression

$$P(D_p) = 0.819 \exp\ (-45.63\Delta) + 0.0976 \exp\ (-89.22\Delta)$$
$$+ 0.035 \exp\ (-228\Delta) + \ldots \qquad (3.5)$$

For small Δ, as more and more terms of Equation (3.5) are needed, Gormley and Kennedy (1949) give an asymptotic solution:

$$P(D_p) = 1 - 6.41\Delta^{2/3} + 4.8\Delta + 1.123\Delta^{4/3} + \ldots \qquad (3.6)$$

Ingham (1975) recomputed the constants and found that the expression given by Gormley and Kennedy (1949) is correct. Thomas (1967) gives an empirical formula:

$$P(D_p) = 0.819 \exp\ (-14.63\Delta) + 0.097 \exp\ (-89.2\Delta)$$
$$+ 0.032 \exp\ (-228\Delta) + 0.027 \exp\ (-492\Delta) \qquad (3.7)$$
$$+ 0.025 \exp\ (-3000\Delta)$$

for all values of Δ. This expression gives reasonable values of Δ down to 1×10^{-6}, and has been used in the present study.

If the CHS holes are assumed to be circular, the penetration of various particles of different sizes through the CHS battery can be calculated using Equation (3.7). Figures 3.7 and 3.8 give the penetration for monodisperse aerosols ranging from 0.0032 to 0.2 um for the CHS battery at flow rates of 4 and 6 lpm. Sinclair et al. (1979) found that experimentally obtained penetration curves for monodisperse aerosols

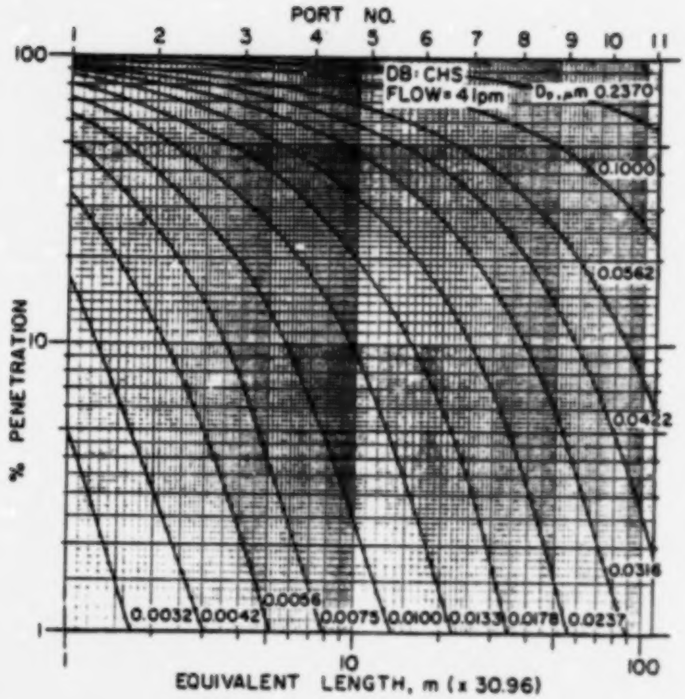

Figure 3.7 Penetration of monodisperse submicron aerosols flowing laminarly in the CHS battery at 4 lpm.

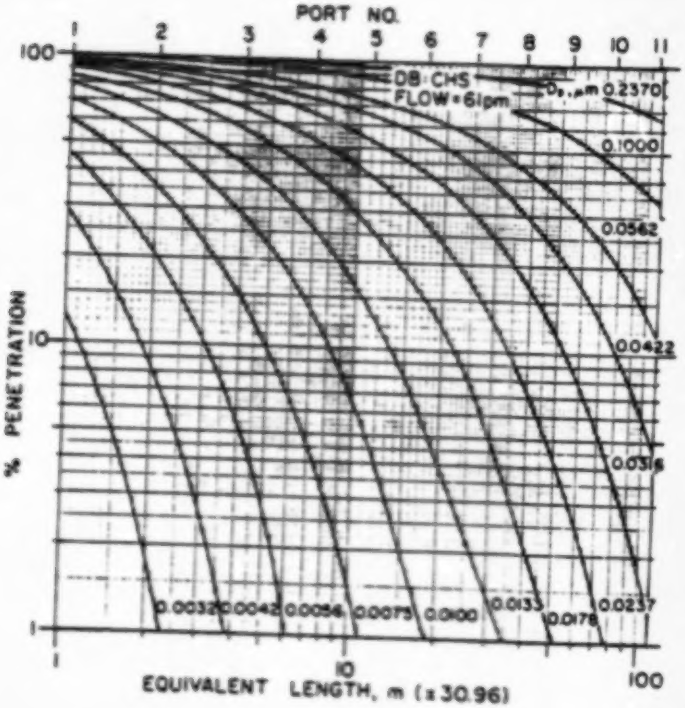

Figure 3.8 Penetration of monodisperse submicron aerosols flowing laminarly in the CHS battery at 6 lpm.

in the size range of 0.01 to 0.1 μm agree closely with the penetration curves obtained by Equation (3.7) for a similar CHS diffusion battery. The theoretical penetration functions have also been verified within experimental error by Thomas (1955) and Kotrappa et al. (1975). The theoretically calculated penetration curves are used in the data reduction procedures described later in this chapter.

For the TSI screen diffusion battery, no adequate theory is available which predicts the diffusion loss in stacks of stainless steel mesh screens. Thus, the response of the battery has to be determined experimentally. The primary calibration of the screen type diffusion battery has been performed by two independent methods by Sinclair and Hoopes (1975) and Sinclair et al. (1979).

In the first method, Sinclair and Hoopes (1975) obtained the penetration curves for the screen and CHS batteries for a polydisperse aerosol. By comparing the two curves, they obtained the equivalent number of screens for each stage of the diffusion battery. They repeated the experiments for three polydisperse aerosols of three different geometric number mean diameters and geometric standard deviations. They found the agreement between the three experiments to be satisfactory. Also, Sinclair et al. (1979) calibrated the screen diffusion battery using monodisperse aerosols generated by the electrical classification method reported by Liu and Pui (1975). Good agreement was found between the calibrations done by the two methods. Figures 3.9 and 3.10 show the penetration of the monodisperse aerosols of several sizes for the TSI screen diffusion battery at flow rates of 4 and 6 lpm. These calibration

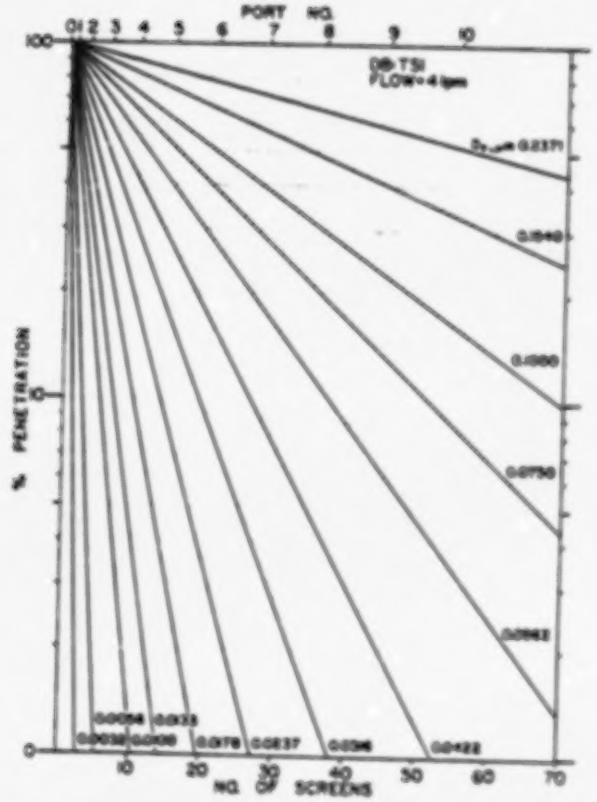

Figure 3.9 Penetration of monodisperse submicron aerosols flowing laminarly in the TSI battery at 4 lpm.

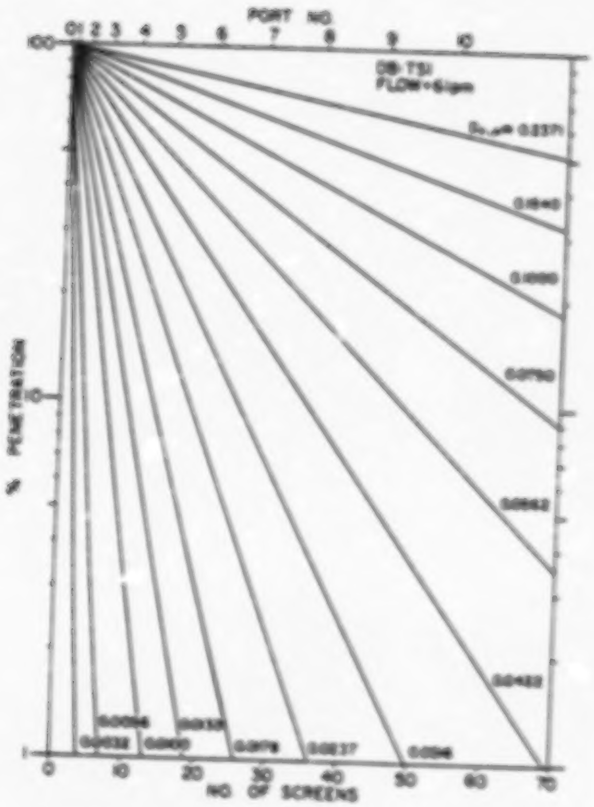

Figure 3.10 Penetration of monodisperse submicron aerosols flowing laminarly in the TSI battery at 6 lpm.

curves were calculated using the procedure described by Sinclair et al. (1979), and are used in the present study to reduce the data taken with the TSI screen diffusion battery.

3.5 Data Reduction

As can be seen from Figures 3.7 through 3.10, monodisperse aerosols of sizes ranging from 0.005 μm to 0.1 μm are lost due to diffusion in various proportions in each of the several stages of the diffusion battery. The diffusion battery can be modeled by Equation (3.8), which is similar to the Fredholm Integral Equation of the First Kind presented in Chapter 2:

$$\Delta L_i = \int S(D_p)\, K_i(D_p)\, f(D_p)\, dD_p + \varepsilon_i \qquad i=1,\ldots I \qquad (3.8)$$

where ΔL_i is the measured diffusion loss in the ith stage, $S(D_p)$ is the sensitivity of the sensor [in this case, the CNC, and as previously explained, $S(D_p)$ is assumed to be 1.0 for all D_p in the present study], $K_i(D_p)$ gives the loss of particles of size D_p in the ith stage, $f(D_p)$ is the size distribution of the aerosol which is to be determined, and ε_i is the error in measurement. The function $K_i(D_p)$ can be determined by taking the difference of penetrations between two consecutive stages, calculated by Equation (3.7). Equation (3.9) is the discretized version of Equation (3.8), obtained in a manner similar to the EAA:

$$\Delta L_i = \sum_{j=1}^{J} K_{ij}\, \Delta N_j + \varepsilon_i \qquad \begin{array}{l} i=1,\ldots I \\ j=1,\ldots J \end{array} \qquad (3.9)$$

Just as in the EAA, a simple inversion of Equation (3.9) with $i=1,\ldots I$ produces a highly oscillating solution, with some of the ΔN_j's being negative. Therefore, several different approaches have been tried to retrieve the size distribution from penetration measurements obtained with diffusion batteries.

3.6 Review of Previous Work

3.6.1 Exhaustion Method (Pollak and Metnieks)

Pollak and Metnieks (1957) used a diffusion battery which consisted of several rectangular channels. They made a set of penetration measurements of aerosols with a Pollak counter by passing the aerosol through the channels at different flow rates. The penetration of particles with diameter D_p flowing through a rectangular channel in a steady stream at a flow rate q, is given by Equation (3.10) (Gormley and Kennedy, 1949):

$$\begin{aligned} \bar{n}/n_0 &= 0.9099 \exp(-x) + 0.053 \exp(-11.4x) + \ldots \\ x &= 3.77\ bLD/qa \end{aligned} \tag{3.10}$$

where D is the diffusion coefficient of particle whose diameter is D_p; L is the length of the diffusion battery; a is the half-width and b is the height of the rectangular channel; n_0 and $\bar{n}$ are the concentrations of the aerosol at the inlet and outlet, respectively; and q is the flow rate through each channel. Neglecting the second term of Equation (3.10) gives:

$$\bar{n}/n_0 = K \exp(-Dy) \tag{3.11}$$

where K = 0.9099, y = 1/fQ, and f = a/3.77bCL, Q = qC, and C = number of channels.

Assuming that the aerosol is made up of m monodisperse fractions and denoting the proportions of constituent i by p_i $(i=1,\ldots m)$, the diffusion coefficient by D_i, and their respective concentrations by n_i, we get:

$$\bar{n} = \sum_{i=1}^{m} \bar{n}_i = \sum_{i=1}^{m} n_{io} K \exp(-y D_i) = n_o K \exp(-y D')$$

or (3.12)

$$\exp(-y D') = \sum_{i=1}^{m} p_i \exp(-y D_i)$$

where D' is the apparent coefficient of diffusion. Pollak and Metnieks showed that, as the flow rate q tends to zero, the apparent diffusion coefficient tends towards the smallest diffusion coefficient in the mixture, and, as q tends to infinity, the apparent diffusion coefficient tends to the average diffusion coefficient of the aerosol.

Thus, the smallest diffusion coefficient, D_1, is obtained by extrapolating the experimental curve of apparent diffusion coefficient vs. flow rate to $Q = 0.0$.

To determine p_i, Pollak and Metnieks rewrote Equation (3.12):

$$\exp(-y D') = p_1 \exp(-y D_1) \quad 1 + \sum_{i=2}^{m} \frac{p_i}{p_1} \exp[-(D'-D_1)y] \qquad (3.13)$$

For small flow rates, and if none of the p_i's are infinitesimal, the term under the summation sign can be neglected, and p_1 is given by Equation (3.14):

$$p_1 = \frac{\exp(-D'\ y)}{\exp(-D_1\ y)} \qquad (3.14)$$

To determine the second component of the aerosol, the contribution due to the first component is subtracted from the experimental curve, and the procedure repeated.

This is done until the final modified apparent coefficient of diffusion is independent of flow rate. The diffusion coefficient of the last component is given by the intersection point of the apparent diffusion coefficient curve with the flow rate axis, and the fraction is given by Equation (3.15):

$$p_m = 1 - \sum_{i=1}^{m-1} p_i \qquad (3.15)$$

This method can be used for aerosols which contain a few (on the order of three) distinct monodisperse components. As most of the aerosols have a continuous size distribution, this method has not been used extensively.

3.6.2 Reverse Method (Fuchs et al.)

Fuchs et al. (1962) proposed a reverse method for reducing the diffusion battery data. In this method, they first calculated the theoretical penetrations of log-normal aerosol size distributions for various values of geometric means and standard deviations in rectangular channels. From these calculations, a set of family of curves of penetration vs. log 1.8852y (where $y = L/h^2 \bar{u}$, L = length of the rectangular channel, h = half-width of the rectangular channel, and $\bar{u}$ = mean linear rate of the flow of the aerosol) were obtained. Experimental values of the penetration of the aerosol size distribution are then plotted on tracing paper against log 1.8852y on the same scale as the first plot. The tracing paper is then placed on the theoretical plot so that the x axis and the corresponding points on them coincide. If the experimental points fall on some theoretical curve, then the parameters of the aerosol size distribution are determined. If the experimental points do not fall on a theoretical curve, the values of the parameters can be obtained by interpolation. If the experimental points do not fall either on a theoretical curve or on a curve obtained by interpolation, then the distribution is obviously not log-normal, and only approximate estimates of the geometrical mean and standard deviation can be obtained. Sansone and Weyel (1971) calculated similar curves for circular tubes. Mercer and Greene (1974) simplified the reverse method by consolidating the number of families of the penetration curves into a single family showing penetration as a function of a median diffusion parameter for several values of standard deviations.

The reverse method of Fuchs being a graphical method, its precision must be regarded with caution, but the underlying concept of fitting a curve to the observed measurements is a good one.

3.6.3 Method of Molan and Scott

Molan and Scott (1963) derived an approximate equation [Equation (3.16)] which gives the equivalent diffusion coefficient, D', as a function of the mean diffusion coefficient, D_0, and the standard deviation of the diffusion coefficient, σ_D,

$$D' = D_0 - \sigma_D^2/2\,k\,q \qquad (3.16)$$

where q is the flow rate through the diffusion battery, and k is the box constant for rectangular channels. The two unknowns, D_0 and σ_D, can be determined from two penetration measurements. This equation is valid only for small σ_D, and thus can be used to reduce diffusion battery data only in special cases.

Metnieks (1965) applied the exhaustion method of Pollak and Metnieks, the reverse method of Fuchs et al., and the method of Nolan and Scott to simulated and experimental diffusion battery data. He found that the exhaustion method and the reverse method were satisfactory for deducing the mean particle size, but none of the three methods gave a satisfactory estimate of the standard deviation or the dispersion of the size distribution.

3.6.4 Inverse Laplace Transform Method (Twomey)

Twomey (1965) proposed yet another technique in which inverse Laplace transforms were used to reduce the diffusion battery data. This method, in principle, can solve the diffusion battery reduction problem, but the penetration measurements have to be known with a precision unattainable either in an experimental or a field measurement situation.

3.6.5 Graphical Stripping Method (Sinclair)

Sinclair (1969) introduced a graphical stripping method to determine the aerosol size distribution from diffusion battery + CNC measurements. This method is similar to Pollak and Metnieks' exhaustion method, in the sense that the aerosol is broken into several monodisperse fractions; however, the size and concentration of each fraction is determined graphically in the stripping method. In the exhaustion method, the largest particle diameter (or smallest diffusion coefficient) is determined by extrapolating the experimental curve to a flow rate of 0.0, and its concentration is calculated by an approximate expression. The graphical stripping procedure is more elegant, and no extrapolations or approximations are needed to determine the fraction of the different monodisperse components. To determine the largest particle size, the slope of the experimental penetration vs. length of diffusion battery is compared with performance curves like Figures 3.7 through 3.10 for large Δ. Similarly, the estimate of the fraction of monodisperse constituent is determined graphically, rather than by an approximate formula, as in the exhaustion method.

After the size and the fraction of the largest constituent is determined, its contribution is subtracted from the experimental data, and the procedure repeated. This is done until only a small residue remains in the experimental curve.

This method is explained in sufficient detail for the practical user in a manual put out by TSI, Inc. for their screen diffusion battery. Also, recently Sinclair et al. (1979) have incorporated this method in a computer program, thus making it easy to use.

Just as in the case of the exhaustion method, the continuous size distribution is divided into a few components in the graphical stripping method. This approximation is valid only if the log of the ratio of the diameter of two consecutive components, i.e., $\log D_i/D_{i+1}$, is small. But this criteria cannot always be met for all experimental data reduced by this method.

3.6.6 Method of Maigne et al.

Maigne et al. (1974) used a diffusion cell made up of rectangular channels. They passed the aerosol through the diffusion cell at several flow rates and made penetration measurements with a condensation nuclei counter. For each flow rate, they calculated a monodisperse particle size which had a penetration of 1% in their diffusion cell. The aerosol size range was then divided into several intervals, with the bounds of the intervals being the particle sizes just calculated. Assuming a uniform rectangular distribution over each interval, they modeled the diffusion battery by a set of linear equations similar to Equation (3.9). By choosing the particle sizes in the manner described above, the coefficient matrix of the linear set of equations is reduced to a lower triangular matrix, in which all the elements above the diagonal are zero. Maigne et al. first solved the diffusion battery data by a straight inversion procedure; however, they obtained a highly oscillatory solution with the concentration of some of the particles becoming negative. To eliminate the oscillations and the negative concentrations, they superimposed on the previous divisions of the aerosol size distribution a coarser set of divisions, with each coarse division containing one or more of the fine divisions. Assuming that over each of the coarse intervals, the aerosol size distribution can be represented by a segment of a log-normal distribution, the set of linear equations is transformed into a set of nonlinear equations with the parameters of log-normal distributions as the variables.

Finally, the set of non-linear equations are solved and concentration in each of the fine divisions determined. This solution does not exhibit any oscillatory behavior; however, there are two implied assumptions in the method just described. Firstly, the assumption of a uniform rectangular aerosol size distribution within the fine divisions is valid only if the size intervals are small. As each size interval corresponds to a penetration measurement at a particular flow rate, a large number of penetration measurements must be made if the data is to be reduced by this method, e.g., Maigne made 18 penetration measurements for an aerosol in the size range 0.018 μm to 0.35 μm. Secondly, though there is a physical basis for assuming that the aerosol size distribution can be modeled by a multi-component log-normal distribution, there is no a priori reason to model the aerosol size distribution by segments of different log-normal distributions over different portions of the particle diameter range.

3.6.7 Method of Soderholm

Soderholm (1975) developed yet another data reduction technique for diffusion battery data. This data reduction technique determines a physical distribution which consists of histograms of N bars. The bars are of equal width on a logarithmic diffusion coefficient scale. The position of the edge of each bar is defined once the maximum and minimum diffusion coefficients of the aerosol are specified. The coefficient matrix of the set of linear equations, similar to Equation (3.9), is calculated under the assumption that the aerosol size distribution is a uniform rectangular one over the interval of each histogram. The set of equations have a total of N+2 parameters, N heights of the histogram, and two variables which specify the edge positions of the aerosol size distribution. The number of parameters are chosen to be less than the number of data points. To solve the set of equations which model the diffusion battery, first the edge parameters are assumed, and the set of equations solved by a linear least squares analysis technique.

The linear least square analysis method gives the heights of the histogram bars and estimates of their standard error. Using a gradient search method, new values of the edge parameters are determined, and the whole procedure repeated until the minimum value of the root mean square difference between the predicted and the measured values relative to the uncertainty in measurements is determined.

Once again, as in the method of Maigne et al., the assumption of a rectangular aerosol size distribution between the bounds of the histogram bar is valid only if the width of the bar is small. Also, the minimum and maximum diffusion coefficient of the aerosol size distribution for data reduction purposes should be determined by the characteristics of the particle sensor used to measure the penetration. Thus, pragmatically, there is no need to vary the edge position parameters.

3.7 Present Work

Knutson (1977) applied Twomey's NLIA, described in Chapter 2, to the diffusion battery data reduction problem. In the present study, Twomey's NLIA has been used to analyze the diffusion battery data. The NLIA(II) described in the second chapter has also been adapted for use with the diffusion battery data. This algorithm has also been used in the present study. Also, a method based on Nelder and Mead's simplex minimization method has been developed and used to analyze the diffusion battery data.

3.7.1 NLIA's Applied to Diffusion Battery

The set of linear equations for the diffusion battery are given by Equation (3.17):

$$\Delta L_i = \sum_{j=1}^{45} S_j R_{ij} \Delta N_j + \epsilon_i \qquad i=1,\ldots I \qquad (3.17)$$

where ΔL_i is the measured diffusion loss in the ith stage, S_j is the sensitivity of the particle sensor, R_{ij} is the response matrix (which is either calculated based on theoretical considerations or determined experimentally), ΔN_j is the particle concentration of the jth size, and ϵ_i is the error associated with each measured ΔL_i. Equation (2.15), when applied to the diffusion battery, gives Equation (3.18):

$$\Delta N_j^{p} = [1 + (\gamma_i^{p-1} - 1) R_{ij}] \Delta N_j^{p-1}$$

with $\qquad (3.18)$

$$\gamma_i^{p-1} = \Delta L_i \quad \sum_{j=1}^{45} S_j R_{ij} \Delta N_j^{p-1} \qquad \begin{matrix} i=1,\ldots I \\ j=1,\ldots 45 \end{matrix}$$

Just as in the EAA, the particle size range from 0.0032 μm to 2.06 μm is divided into 45 equal geometric divisions. The subscript j in Equation (3.18) corresponds to the 45 divisions in the particle diameter range, and the subscript i corresponds to the measured diffusion losses in the stages of the diffusion battery, with i=1 corresponding to the penetration measured at the end of the last stage. For the CHS battery, I has a value of 12 (11 stages), and for the screen diffusion battery, I takes on a value of 11 (10 stages).

To start the iterative procedure, a starting solution as given by Knutson (1977) has been used in the present study.

$$\Delta N_j^{1} = \sum_{i=1}^{I} \Delta L_i \quad 45 \qquad j=1,\ldots 45 \qquad (3.19)$$

Equation (3.19) gives a uniform aerosol size distribution as the starting solution. For the convergence criteria, Knutson (1977) found that after

30 iterations, the change in size distributions between two consecutive iterations was negligible, and thus terminated the iterative procedure after 30 iterations. In the present study, at first 20 iterations are carried out; then WLS according to Equation (3.20) is computed.

$$WLS = \sum_{i=1}^{I} \left(\frac{\Delta L_i'}{\sum_{k=1}^{I} \Delta L_k'} - \frac{\Delta L_i}{\sum_{k=1}^{I} \Delta L_k} \right)^2 \Big/ \frac{\Delta L_i}{\sum_{k=1}^{I} \Delta L_k} \qquad (3.20)$$

where $\Delta L_i'$ is the predicted loss in the ith stage according to Equation (3.17). If the WLS is less than 0.005, the iterative process is stopped; otherwise, more iterations are done in increments of 5, until the desired value of WLS is reached. After a maximum of 100 iterations, the iterative procedure is halted even though WLS is not below the set value of 0.005.

When the iterative process has been stopped, a set of 45 values of ΔN_j's are determined which fit the data acceptably. Just as in the EAA, the ΔN_j's are then averaged over an interval of diameter such that 10 histogram bars are obtained. The bounds of the histogram were chosen so as to coincide with the channel boundaries of the EAA, so that direct comparison can be made easily between the measured distributions by the EAA and the diffusion battery method.

Similarly, Equation (2.24), when applied to the diffusion battery, gives:

$$\Delta N_j^{p} = \Delta N_j^{p-1} \times \sum_{i=1}^{I} R_{ij} \, \gamma_i^{p-1} \qquad j=1,\ldots 45 \qquad (3.21)$$

The starting solution, the stopping criteria, and the final presentation of the data are done exactly in the same manner as for Twomey's NLIA.

3.7.1.1 Simulation Studies for NLIA

Just as for the EAA, simulation studies for the diffusion battery were done in two stages. In the first stage, the response of the CHS battery (at a flow rate of 4 lpm) was calculated for several unimodal log-normal distributions. The values of the parameters for the unimodal log-normal distributions are given in Table 3.2. The calculated responses for these distributions were used as input to the NLIA's. Figure 3.11 shows the variation of WLS with the number of iterations for some of the simulation runs. Here again, the WLS drops rapidly initially, but the change of WLS is very small as the number of iterations increase, both for Twomey's NLIA and for NLIA(II). Figures 3.12(a) and (b) show that the retrieved histogram is very close to the input distribution for both Twomey's NLIA and for NLIA(II). This was observed for all simulation runs done with the diffusion battery. An additional point to be noted here is that the retrieved distribution does not oscillate, as was the case with the EAA. This can be attributed to the fact that for the diffusion battery, the penetration curves are not as sharp as the current vs. voltage curves for the EAA, and do not exhibit discontinuities as they do in the EAA.

For the second stage, a normal random error (mean 0, and a of 5% of ΔL_i, $i=1,\ldots 11$) was added to ΔL_i, $i=1,\ldots 11$ (where ΔL_i is the calculated diffusion loss in the i^{th} stage of the battery for an input aerosol size distribution). These modified sets of ΔL_i's were then used as inputs to the NLIA's, and their solutions determined.

Table 3.2 Log-normal distributions used for diffusion battery (CHS, flow = 4 lpm) simulation studies for NLIA's

Distribution No.	Parameters	
	DGN μm	σ_g
1	0.01	1.3
2	0.01	2.0
3	0.03	1.3
4	0.03	2.0
5	0.05	1.3
6	0.05	2.0
7	0.07	1.3
8	0.07	2.0

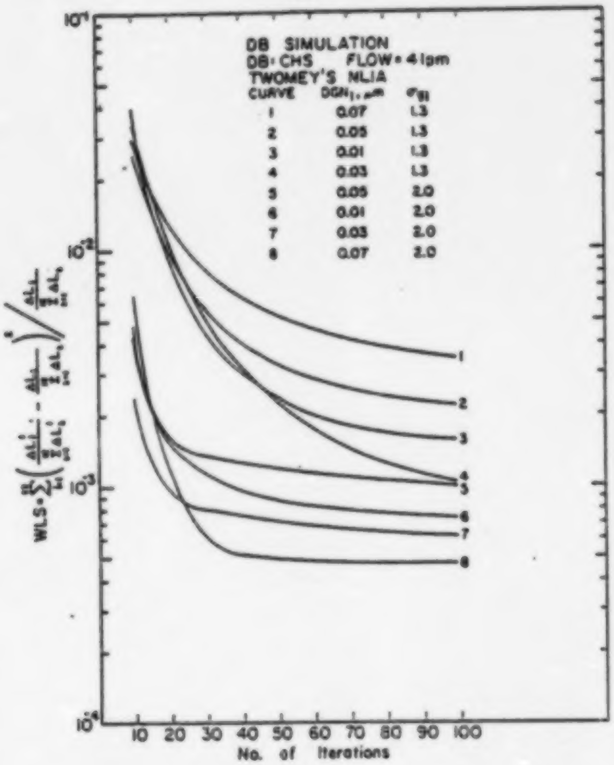

Figure 3.11(a) WLS vs. number of iterations for CHS battery (4 lpm) simulated data, using Twomey's NLIA data reduction method.

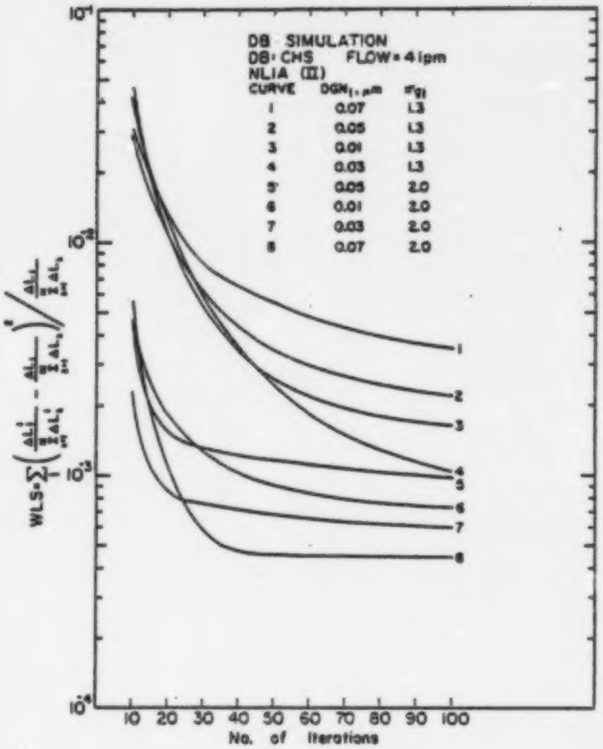

Figure 3.11(b) WLS vs. number of iterations for CHS battery (4 lpm) simulated data, using NLIA(II) data reduction method.

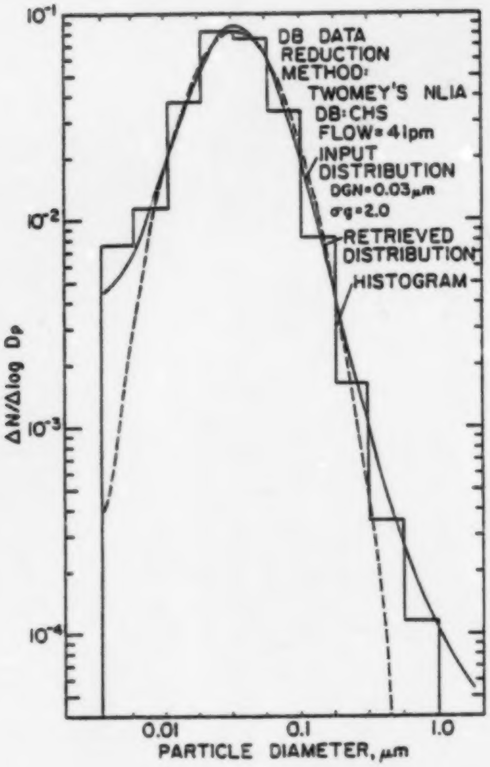

Figure 3.12(a) Retrieved number distribution using Twomey's NLIA for a simulated response of the diffusion battery + CNC to a unimodal log-normal distribution (DGN = 0.03 μm, σ_g = 2.0).

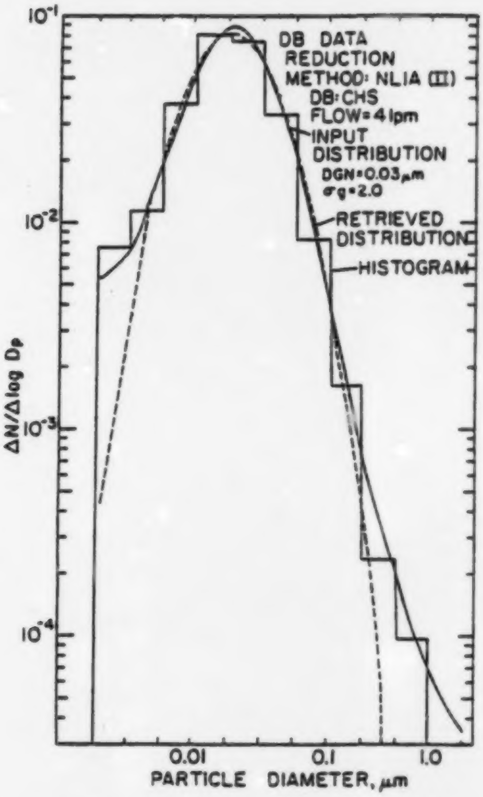

Figure 3.12(b) Retrieved number distribution using NLIA(II) for a simulated response of the diffusion battery + CNC to a unimodal log-normal distribution (DGN = 0.03 μm, σ_g = 2.0).

Again, just as for the EAA, the results of the second part of the study agreed to within ±10% of those obtained in the first part.

3.7.2 Simplex Minimization Method Applied to the Diffusion Battery

As for the EAA, a normalized bimodal log-normal distribution given by Equation (2.31) is used to model the aerosol size distribution. The objective function used for this method is given by Equation (3.13). Using the simplex minimization algorithm, the optimum set of parameters which best fit the data are obtained.

To start the simplex minimization procedure, initial estimates of the parameters are used as an input. The initial estimates do not have to be very accurate, but the closer the initial estimates are to the optimum set of parameters, the less is the computing cost. Just as for the EAA, the final value of WLS can be used as a flag to determine whether the final set of optimum parameters do indeed represent the global minimum or are very close to it. Generally, a value of 0.03 is obtained for the final value of WLS. If the final value of WLS is large, the simplex minimization procedure must be restarted using a different set of initial estimates.

For estimating the standard errors of the parameters of the optimum log-normal distribution, the square root of the diagonal elements of the Hessian are calculated in a manner similar to that for the EAA. The proportionality constant for the standard error of measurement was assumed to be 7.5% of the square root of the normalized loss for the laboratory-generated aerosols.

3.7.2.1 Simulation Studies for Simplex Minimization Method

To determine the applicability of the simplex minimization method to the diffusion battery reduction problem, a set of simulation runs similar to the EAA were made. Random normal noise with a standard deviation of 5%, 10%, and 15% of the calculated diffusion loss was added to the calculated response of the diffusion battery. The results of the simulation runs are summarized in Table 3.3. The simplex minimization method can retrieve the geometric number mean diameter from diffusion battery data satisfactorily, but the discrepancy between the retrieved estimates and the input values of the geometric standard deviation increases as the number mean diameter increases. In Chapter 4, it is shown that this is not an artifact of the data reduction procedure, but is a direct consequence of the diffusion battery method itself.

3.8 Summary

In this chapter, the diffusion battery method of measuring the aerosol size distribution was discussed. Two diffusion batteries, the CHS and the TSI screen battery, were described, together with the TSI continuous condensation nucleus counter, which was used in the present study as the particle sensor. A brief statement of the data reduction problem, with a review of the various methods developed by other investigators, was also presented. Two new data reduction methods developed in the course of this thesis were also discussed. The computer programs for the data reduction methods developed and analyzed in the present thesis are presented in the appendices.

Table 3.3 Comparison of the input and corresponding retrieved distribution parameters of log-normal aerosol distributions for simplex minimization method simulation studies for the DB

Case	DGN (μm)	σg	WLS
A[1]	0.0100	2.00	---
B[2]	0.0111	1.79	4.030×10^{-3}
C[3]	0.0110	1.76	8.000×10^{-3}
D[4]	0.0107	1.71	1.403×10^{-2}
A	0.0100	1.30	---
B	0 0102	1.21	2.143×10^{-3}
C	0.0103	1.21	4.574×10^{-3}
D	0.0105	1.16	9.195×10^{-3}
A	0.0300	1.03	---
B	0.0290	2.05	4.940×10^{-3}
C	0.0288	1.92	9.084×10^{-3}
D	0.0288	1.90	1.491×10^{-2}
A	0.0300	1.30	---
B	0.0304	1.30	2.971×10^{-3}
C	0.0311	1.26	9.072×10^{-3}
D	0.0318	1.25	2.040×10^{-2}

(continued)

[1] A: Input distribution

[2] B: Retrieved distribution for 5% error case

[3] C: Retrieved distribution for 10% error case

[4] D: Retrieved distribution for 15% error case

Table 3.3 (continued)

Case	DGN (μm)	σ_g	WLS
A	0.0500	2.00	---
B	0.0487	1.96	3.627×10^{-3}
C	0.0466	1.85	8.296×10^{-3}
D	0.0468	1.73	1.639×10^{-2}
A	0.0500	1.30	---
B	0.0502	1.34	1.592×10^{-3}
C	0.0515	1.26	3.165×10^{-3}
D	0.0515	1.26	6.890×10^{-3}
A	0.0700	2.00	---
B	0.0700	2.00	1.068×10^{-3}
C	0.0680	1.97	3.827×10^{-3}
D	0.0676	1.67	8.872×10^{-3}
A	0.0700	1.30	---
B	0.0690	1.37	5.739×10^{-3}
C	0.0683	1.45	1.213×10^{-2}
D	0.0681	1.51	2.204×10^{-2}

CHAPTER 4

EXPERIMENTAL STUDY

4.1 Introduction

Although the electrical aerosol analyzer and the diffusion battery have been in use to measure submicron aerosols for some time, an extensive comparison of the two methods has been lacking. The only previous work in this area has been performed by Bricard et al. (1977). In their work, Bricard et al. used an aerosol produced by radiolysis of gaseous impurities of previously filtered atmospheric air. They found that both the EAA and the diffusion battery give reasonable estimates of the number mean diameters of aerosol size distributions for DGN greater than 0.02 μm; however, no reasonable comparison of the estimates of geometric standard deviation by the two methods is available.

In the present study, submicron aerosols of di-octyl phthalate (DOP) and NaCl were generated using a Collison atomizer. These aerosols were then simultaneously measured by the EAA and the diffusion battery. The data thus obtained were analyzed using the data reduction methods described previously and compared.

4.2 Experimental Set-Up

A schematic of the system for measuring the size distribution of aerosols using the EAA and the diffusion battery+CNC is shown in Figure 4.1(a). The system consists of an aerosol generator, a diffusion type silica gel dryer, a Kr-85 neutralizer, an EAA, the diffusion batteries, the TSI continuous flow condensation nucleus counter, a pump,

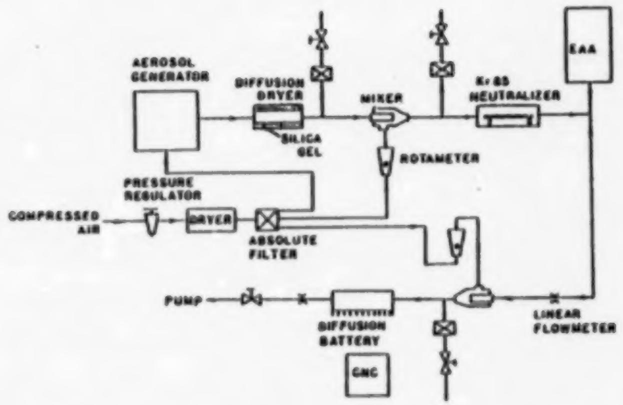
AEROSOL GENERATOR
DIFFUSION DRYER
SILICA GEL
MIXER
Kr 85 NEUTRALIZER
EAA
ROTAMETER
PRESSURE REGULATOR
COMPRESSED AIR
DRYER
ABSOLUTE FILTER
PUMP
DIFFUSION BATTERY
CNC
LINEAR FLOWMETER

Figure 4.1 (a) Schematic of the experimental set-up for measuring simultaneously the size distribution of aerosols with the EAA and diffusion battery + CNC.

diluters, and capillary tube type linear flow meters. The EAA, the diffusion batteries, and the CNC have been described in Chapters 2 and 3. The aerosol generator is described below.

4.2.1 Aerosol Generator

The aerosol generator used for the experiments is based on the one developed by Liu and Lee (1975) and further modified by Rubow (1979). The generator is essentially a Collison atomizer with separate reservoirs for feed and waste liquids. In the Liu and Lee design, fresh solution was fed to the atomizer by a syringe pump and the waste solution collected in a separate bottle. The long term average output of the aerosol was fairly stable, but for the short term, it showed a cyclical variation of ±10% in concentration with a period of approximately three to four minutes.

Rubow replaced the syringe pump with a capillary tube feed system, thus avoiding the cyclical variations of the gear-driven syringe pump. Figure 4.1(b) shows the schematic of the generator. The high velocity jet downstream of the orifice produces a drop in the pressure, which provides the driving force to aspirate the liquid in the atomizer. To control the flow rate of the fresh solution, a restriction in the form of a capillary tube is placed in the path of the fresh liquid. Rubow used a stainless steel capillary tube 2.5 cm long with a 0.025 cm ID. The liquid flow rate was determined by him to be 0.57 cc/min for alcohol and a waste liquid accumulation rate of 0.28 cc/min. Rubow also modified the outlet of the aerosol generator to eliminate the liquid build-up on the top horizontal wall of the atomizer. To reduce the polydispersity

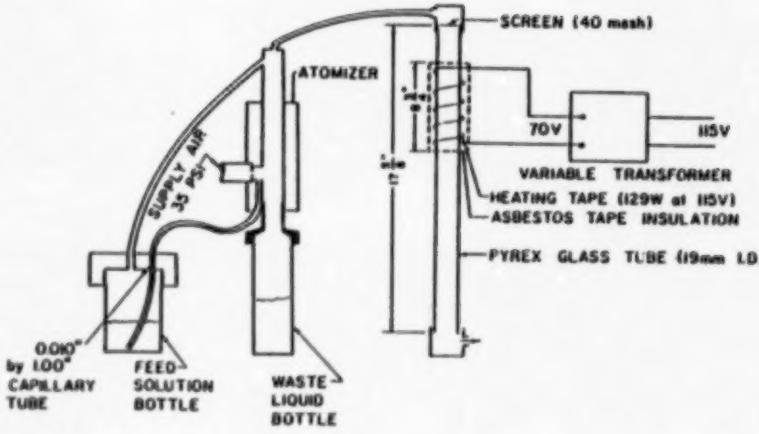
SCREEN (40 mesh)
ATOMIZER
8 3/4"
17 3/8"
70V
115V
VARIABLE TRANSFORMER
HEATING TAPE (129W at 115V)
ASBESTOS TAPE INSULATION
PYREX GLASS TUBE (19mm I.D.)
SUPPLY AIR 35 PSI
0.010" by 1.00" CAPILLARY TUBE
FEED SOLUTION BOTTLE
WASTE LIQUID BOTTLE

Figure 4.1 (b) Schematic of the aerosol generator.

of the atomized aerosol, the output from the Collison atomizer is passed through a vaporizer/condensor tube. This is a simplified version of the condensor generator developed by Liu et al. (1966).

4.2.2 Experimental Procedure

As mentioned previously, aerosols of two different materials (NaCl and DOP) were used in the present study. The number mean diameters of the aerosol were varied by using solutions of different concentrations (0.01% NaCl, 0.01% DOP, 0.037% DOP, and 0.1% DOP). For the DOP aerosols, the vaporizer/condensor tube was used; however, for the NaCl aerosol, it was bypassed. Thus, the DOP aerosols had different geometric number mean diameters with relatively the same dispersion, while the NaCl aerosol had a different geometric number mean diameter and geometric standard deviation.

For each aerosol, four diffusion battery measurements were made (CMS and TSI screen diffusion batteries, at two flow rates each: 4.0 and 6.0 lpm), and between six to eight measurements with the EAA. To insure that the aerosol was stable while the diffusion battery measurements were being made, the total current from the EAA was monitored on a strip chart recorder.

The atomizer described previously generates a unimodal aerosol. A bimodal aerosol can be obtained by mixing the output of two such atomizers which use feed solutions of different concentrations. As both the EAA and the diffusion battery can be modeled by sets of linear equations [Equations (2.8) and (3.17)], data for bimodal aerosol size distribution can be obtained by summing the response of two unimodal aerosols for each of the instruments.

For the present study, bimodal aerosol data for the EAA was obtained by adding a fraction of the response of the EAA to NaCl aerosol to the currents measured with the DOP aerosols. The value of the fraction was chosen such that the number fraction in the first mode was 0.5. Similarly, for the diffusion battery, the penetration of NaCl aerosol was added to the penetration of DOP aerosols for CHS battery to obtain data for bimodal aerosols.

4.3 Results

The EAA data was reduced using Twomey's NLIA, NLIA(II), the simplex minimization technique, and the monodisperse sensitivity method of Liu and Pui (1975). Similarly, the diffusion battery data was reduced using Twomey's NLIA, NLIA(II), and the simplex minimization technique. In the following paragraphs, some of the results are presented.

4.3.1 Reduced EAA Data

Figures 4.2 to 4.5 give the histograms of the aerosol size distribution as measured with the EAA, using the different data reduction techniques. Table 4.1 gives the estimates of the total number, surface, and volume concentrations obtained with the different data reduction methods. The first column in Table 4.2 gives the estimated geometric number mean diameter, the geometric standard deviation, and their standard errors of estimation, as retrieved by the simplex minimization method.

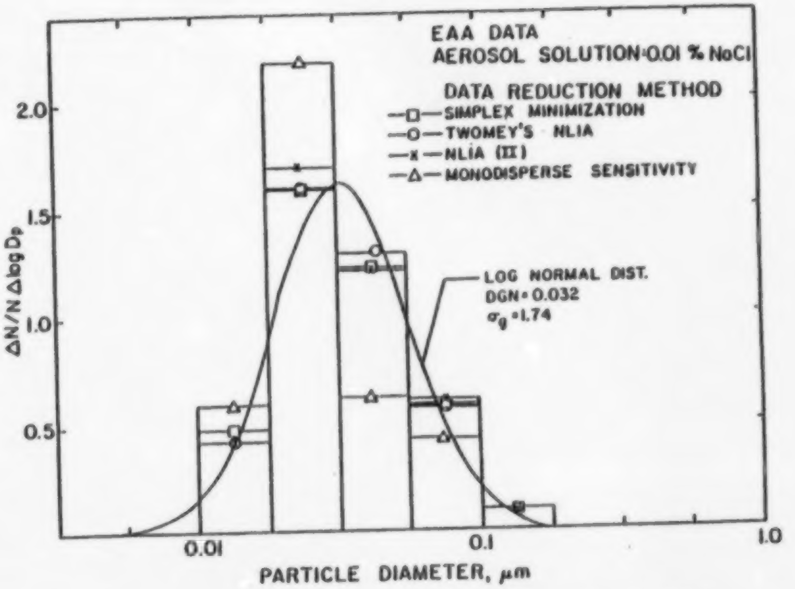

Figure 4.2(a) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl solution): number

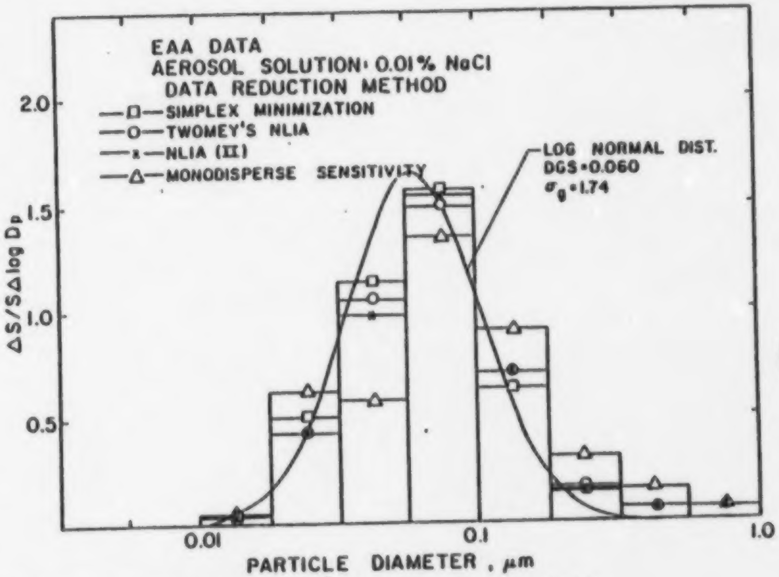

Figure 4.2(b) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl solution): surface

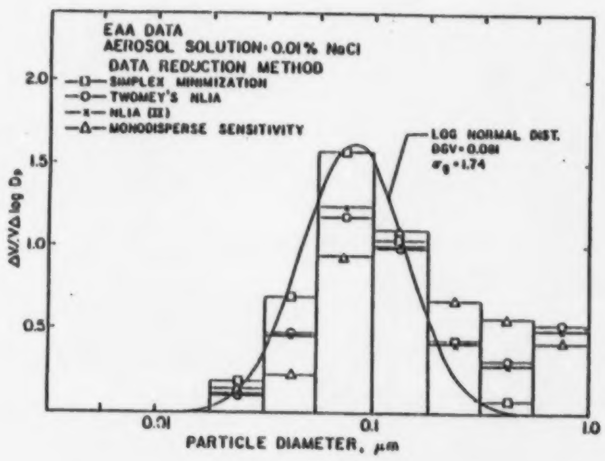

Figure 4.2(c) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl solution): volume

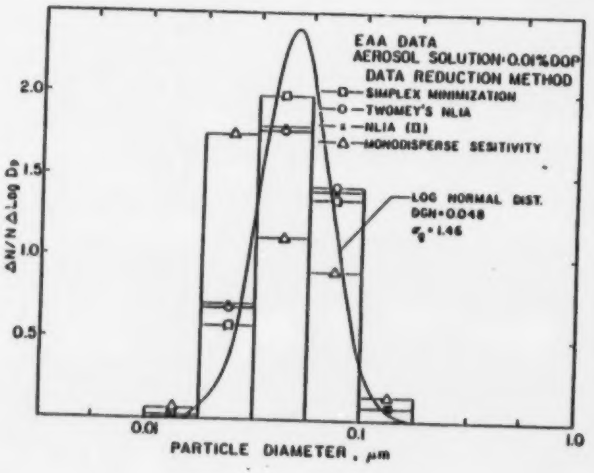

Figure 4.3(a) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% DOP solution aerosol): number

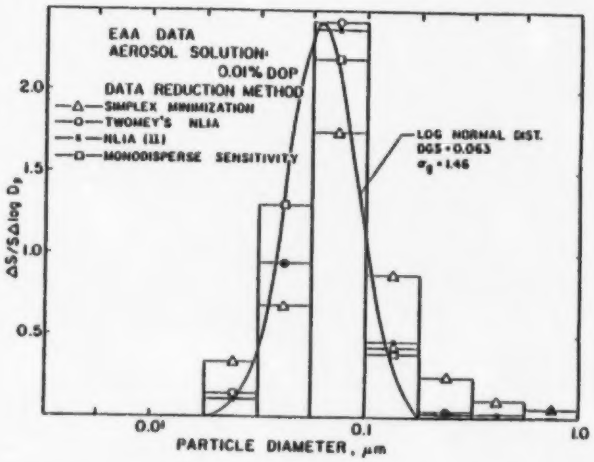

Figure 4.3(b) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% DOP solution aerosol): surface

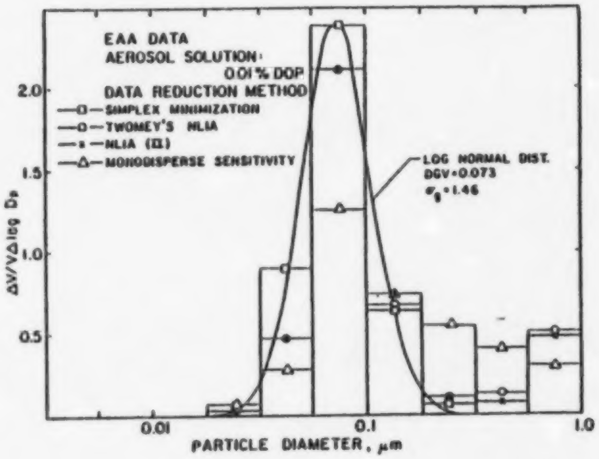

Figure 4.3(c) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% DOP solution aerosol); volume

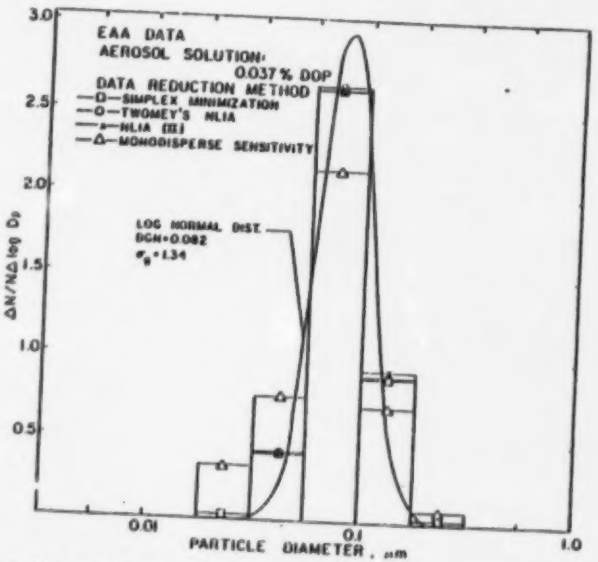

Figure 4.4(a) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.03% DOP solution aerosol): number

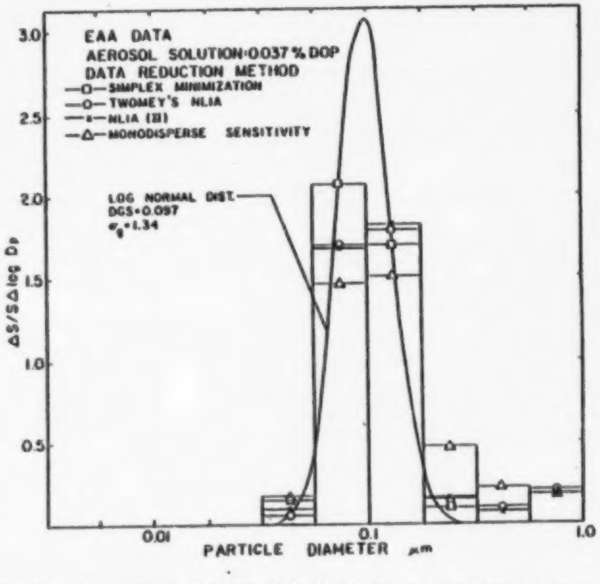

Figure 4.4(b) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.03% DOP solution aerosol): surface

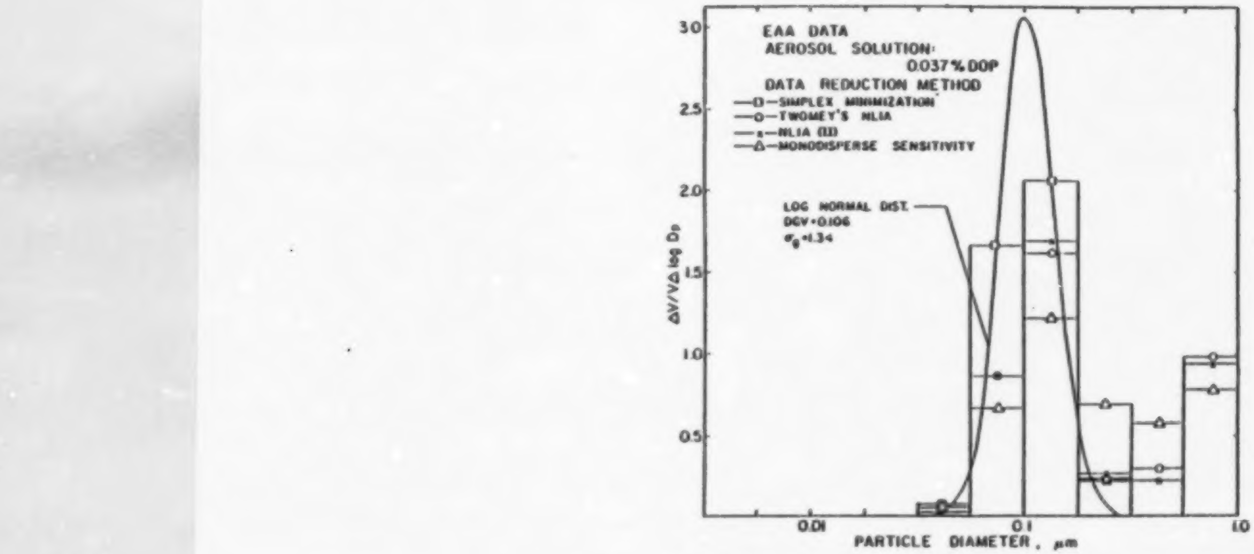

Figure 4.4(c) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.03% DOP solution aerosol): volume

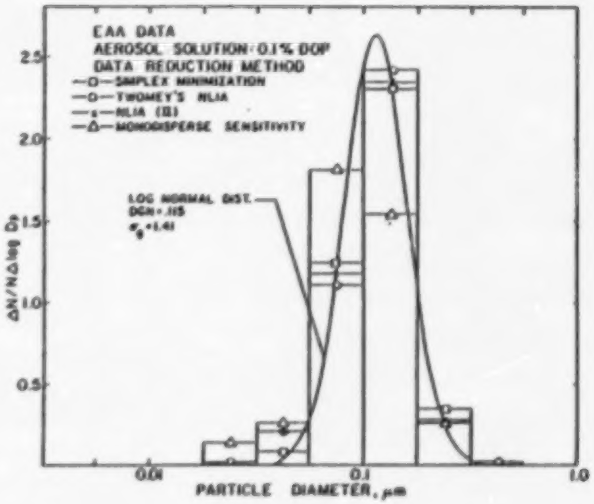

Figure 4.5(a) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.1% DOP solution aerosol): number

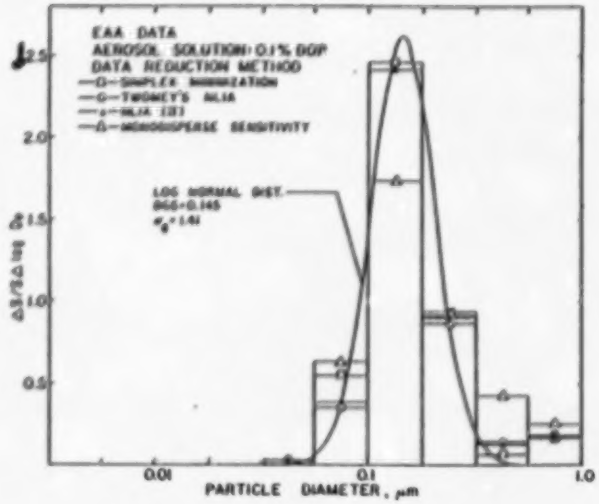

Figure 4.5(b) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.1% DOP solution aerosol): surface

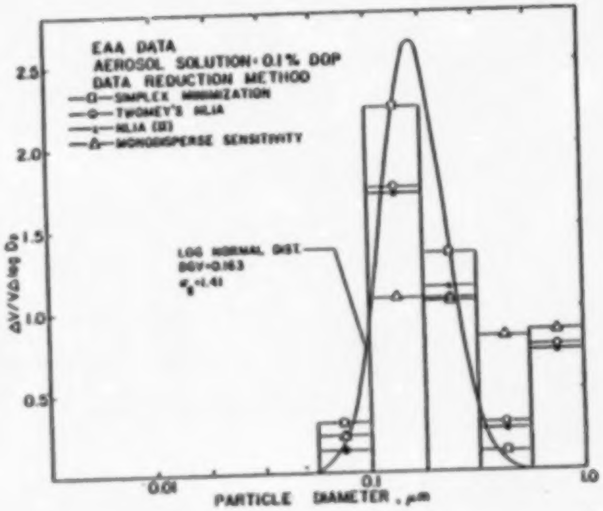

Figure 4.5(c) Histogram of the normalized aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.1% DOP solution aerosol): volume

Table 4.1 Comparison of total concentration estimates for the EAA data reduction method

Data Reduction Method	MS[1]	TNLIA[2]	NLIA(II)[3]	SMCFM[4]
Aerosol	Number Concentration (part/cm^3)			
0.01% NaCl	2.021×10^5	1.931×10^5	1.936×10^5	1.853×10^5
0.01% DOP	6.568×10^4	6.179×10^4	6.196×10^4	6.143×10^4
0.037% DOP	5.572×10^4	5.329×10^4	5.318×10^4	5.313×10^4
0.1% DOP	5.446×10^4	4.945×10^4	4.945×10^4	4.998×10^4
	Surface Concentration (μm^2/cc)			
0.01% NaCl	1.247×10^3	1.323×10^3	1.322×10^3	1.170×10^3
0.01% DOP	6.104×10^2	6.507×10^2	6.473×10^2	5.863×10^2
0.037% DOP	1.445×10^3	1.477×10^3	1.471×10^3	1.317×10^3
0.1% DOP	2.681×10^3	2.704×10^3	2.691×10^3	2.584×10^3
	Volume Concentration (μm^3/cc)			
0.01% NaCl	2.278×10^1	2.078×10^1	2.053×10^1	1.434×10^1
0.01% DOP	1.060×10^1	9.224	9.031	6.620
0.037% DOP	3.922×10^1	3.614×10^1	3.525×10^1	2.227×10^1
0.1% DOP	9.549×10^1	8.500×10^1	8.405×10^1	6.528×10^1

1) Monodisperse sensitivity method

2) Twomey's non-linear iterative algorithm

3) Non-linear iterative algorithm (II)

4) Simplex minimization curve fitting method

Table 4.2 Parameters of unimodal log-normal number distribution determined using the simplex minimization curve-fitting procedure

Aerosol Solution	EAA	[illegible] CPC Flow: 0.3 lpm	[illegible] CPC Flow: 4 lpm	[illegible] TSI Flow: 0.3 lpm	[illegible] TSI Flow: 4 lpm
0.015 NaCl in water					
NMD (µm)	0.0205 (0.001)*	0.0169 (0.001)	0.0193 (0.002)	0.0167 (0.001)	0.0158 (0.002)
σ_g	1.75 (0.03)	1.73 (0.19)	1.64 (0.16)	1.60 (0.21)	1.63 (0.26)
WLS	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
0.010 DOP in isopropyl alcohol					
NMD (µm)	0.0415 (0.001)	0.0442 (0.002)	0.0409 (0.002)	0.0442 (0.003)	0.0359 (0.003)
σ_g	1.46 (0.06)	1.37 (0.21)	1.36 (0.19)	1.38 (0.22)	1.33 (0.24)
WLS	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
0.0015 DOP in isopropyl alcohol					
NMD (µm)	0.0617 (0.002)	0.0538 (0.004)	0.0539 (0.004)	0.0736 (0.004)	0.0723 (0.003)
σ_g	1.39 (0.05)	1.46 (0.35)	1.46 (0.36)	1.25 (0.37)	1.38 (0.34)
WLS	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
0.15 DOP in isopropyl alcohol					
NMD (µm)	0.1160 (0.001)	0.103 (0.006)	0.103 (0.006)	0.073 (0.004)	0.0988 (0.003)
σ_g	1.45 (0.03)	1.39 (0.50)	1.46 (0.3)	1.16 (0.75)	1.65 (0.35)
WLS	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

* Standard error estimate

Figures 4.6 to 4.9 give the histogram of the composite aerosol size distribution obtained by summing the response of the EAA for two unimodal aerosols. Table 4.3 gives the estimates of the log-normal number distribution for the bimodal data obtained by using the simplex minimization curve fitting procedure.

4.3.2 Reduced Diffusion Battery-CNC Data

Figures 4.10 to 4.13 give the histograms of the aerosol number size distribution measured with the diffusion battery+CNC, using the different data reduction techniques. The last four columns of Table 4.2 give the estimated geometric number mean diameter, the geometric standard deviation, and their standard errors of estimation as retrieved by the simplex minimization method.

Figures 4.14 to 4.16 give the histograms of the bimodal aerosol, and the parameters of the log-normal number size distribution are given in Table 4.3.

4.4 Discussion

In the following paragraphs, first a comparison of the various data reduction techniques for the EAA is presented, followed by a comparison of the data reduction techniques for the diffusion battery+CNC. As the same data is analyzed using different methods, the differences in the final aerosol size distribution can be attributed solely to the data reduction methods. Finally, a comparison of the aerosol size distribution measuring capabilities of the two instruments, i.e. the EAA and the diffusion battery+CNC, is also presented.

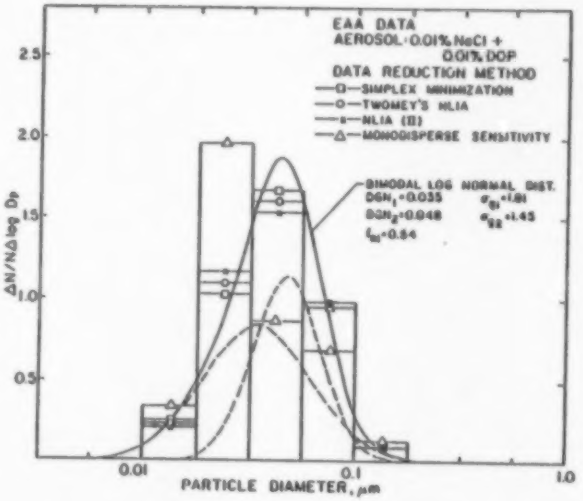

Figure 4.6(a) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by the simplex minimization data reduction method (0.01% NaCl + 0.037% DOP): number

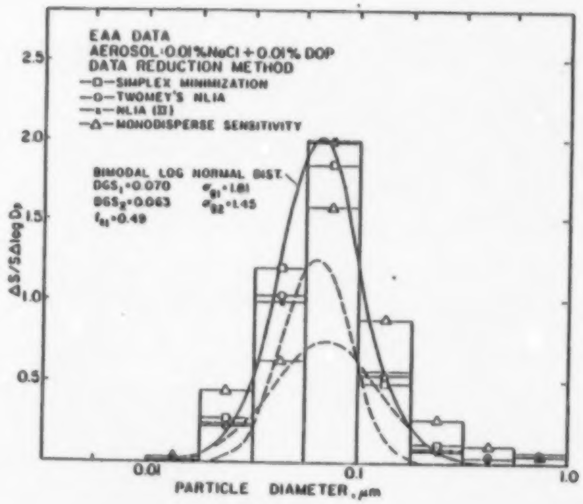

Figure 4.6(b) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by the simplex minimization data reduction method (0.01% NaCl + 0.037% DOP): surface

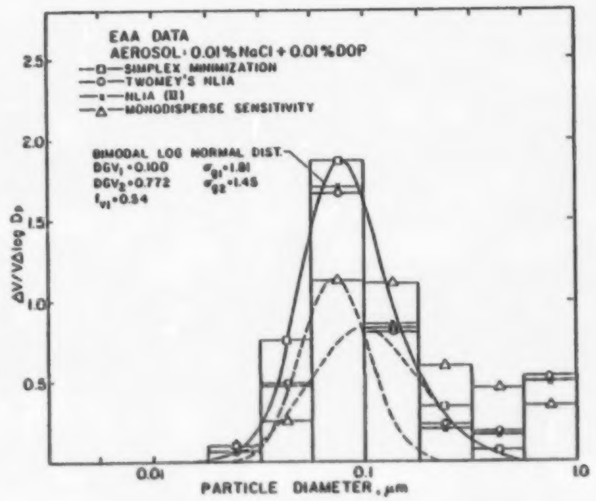

Figure 4.6(c) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by the simplex minimization data reduction method (0.01% NaCl + 0.037% DOP): volume

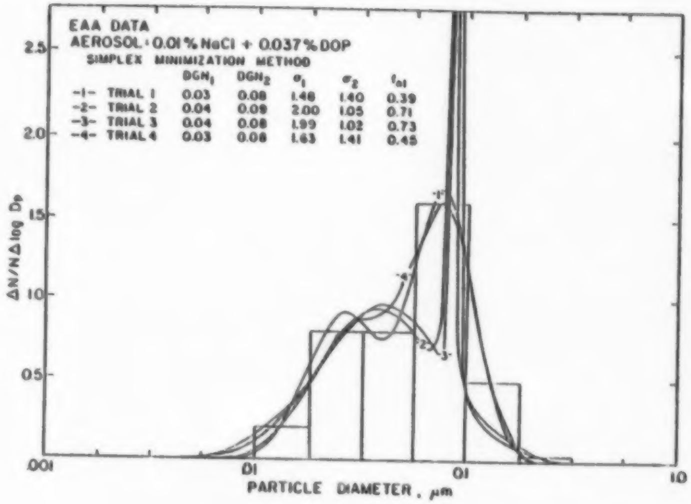

Figure 4.7(a) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.037% DOP): number

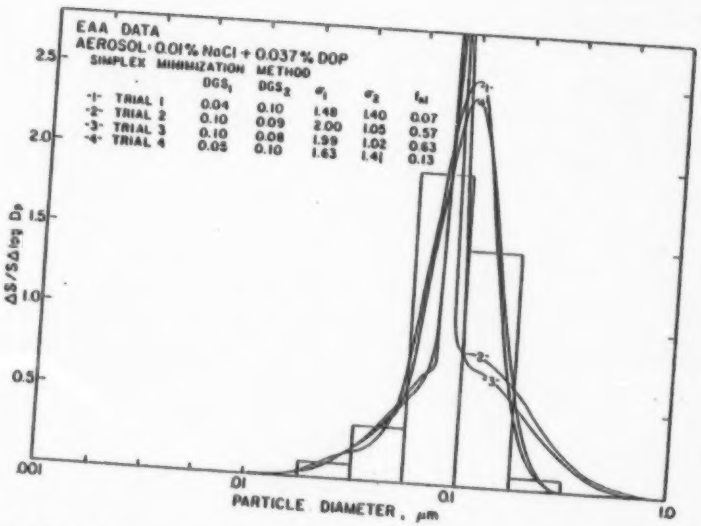

Figure 4.7(b) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.037% DOP): surface

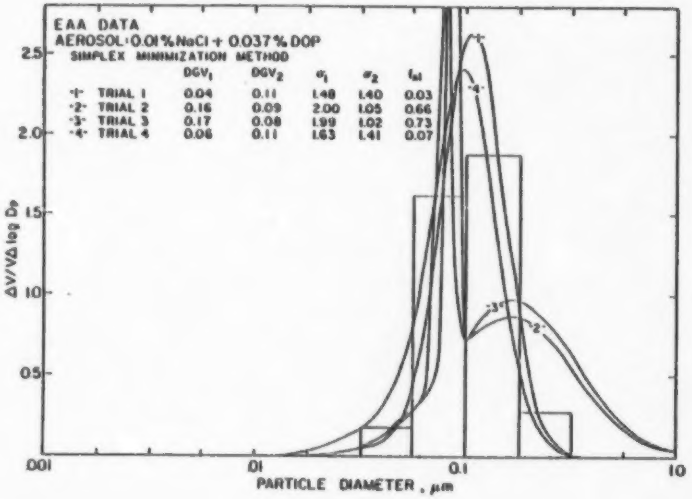

Figure 4.7(c) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.037% DOP): volume

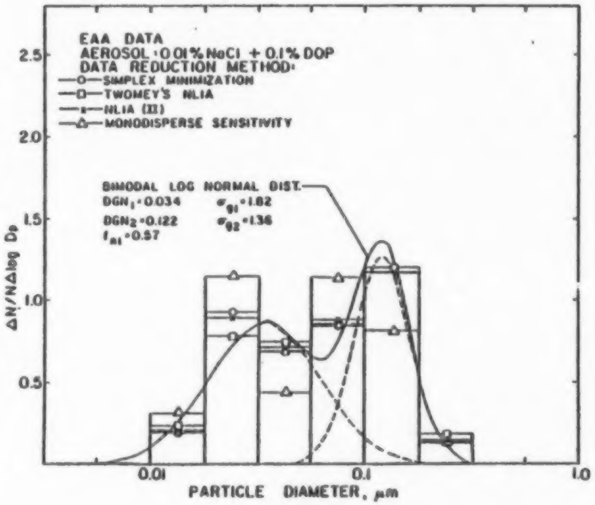

Figure 4.8(a) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.1% DOP): number

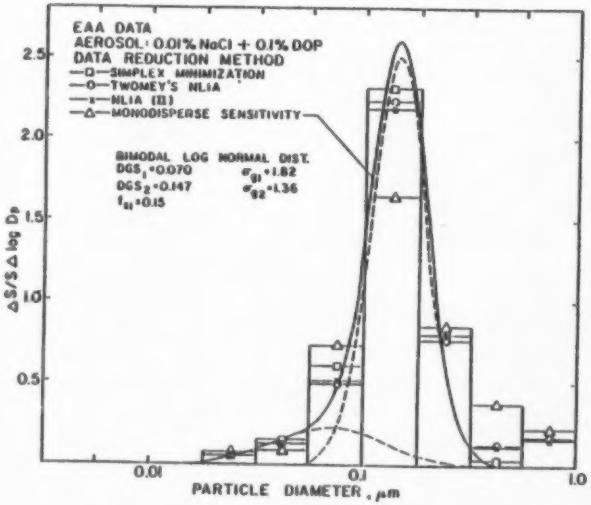

Figure 4.8(b) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.1% DOP): surface

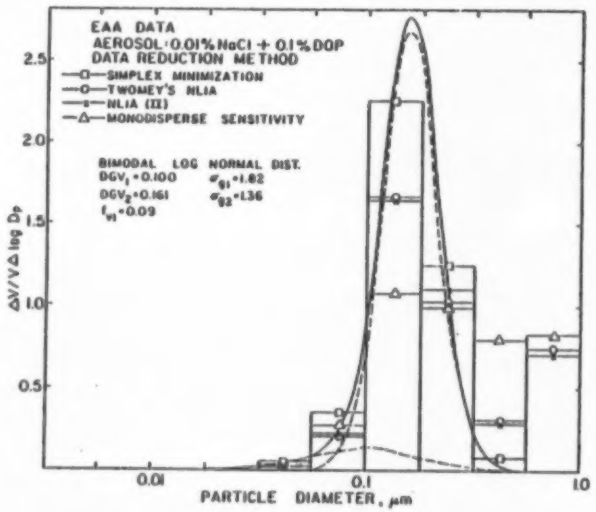

Figure 4.8(c) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.1% DOP): volume

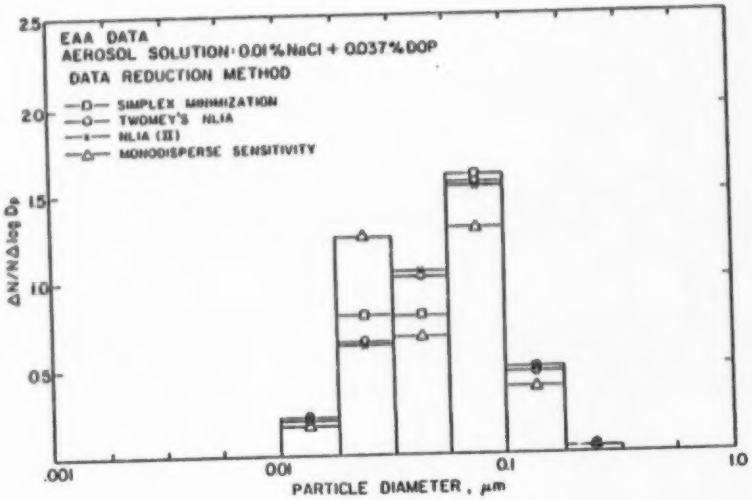

Figure 4.9(a) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.037% DOP): number

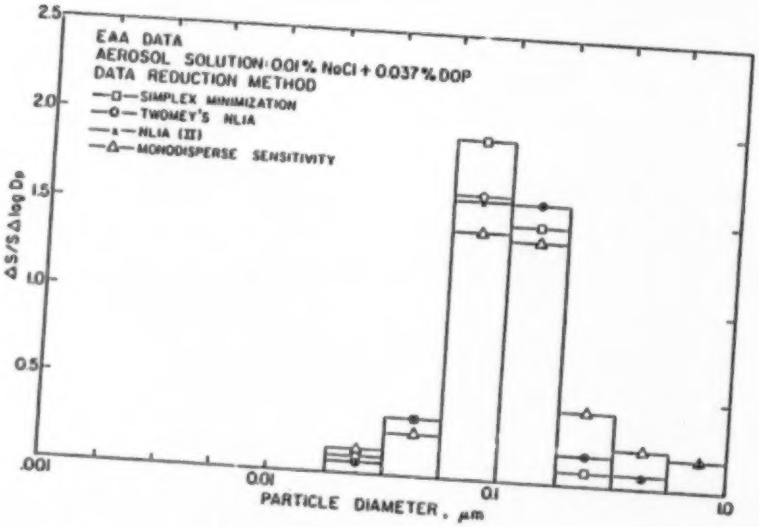

Figure 4.9(b) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.037% DOP): surface

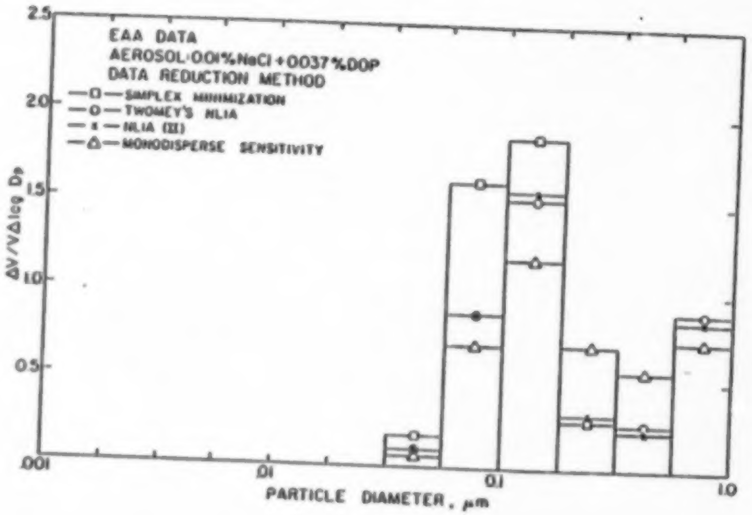

Figure 4.9(c) Histogram of the normalized composite aerosol size distribution measured with the EAA, obtained by different data reduction techniques (0.01% NaCl + 0.037% DOP): volume

Table 4.3 Parameters of bimodal log-normal number distributions determined using the simplex minimization curve fitting procedure

Table 4.3(a)

	EAA	DB
0.01% NaCl + 0.01% DOP		
DGN_1	0.0346	0.0336
σ_1	1.81	1.77
DGN_2	0.0447	0.0462
σ_2	1.45	1.20
f_{n_1}	0.54	0.56
WLS	1.375×10^{-2}	2.972×10^{-3}
0.01% NaCl + 0.037% DOP		
DGN_1	See Table 4.3(b)	0.0353
σ_1		1.85
DGN_2		0.0746
σ_2		1.11
f_{n_1}		0.49
WLS		9.827×10^{-3}
0.01% NaCl + 0.1% DOP		
DGN_1	0.0340	0.0347
σ_1	1.82	2.01
DGN_2	0.1215	0.0945
σ_2	1.36	1.13
f_{n_1}	0.57	0.53
WLS	1.566×10^{-2}	3.021×10^{-3}

Table 4.3(b)

	0.01% NaCl + 0.037% DOP					
EAA Data	DGM_1	σ_1	DGM_2	σ_2	f_{m_1}	WLS
Trial 1	0.0262	1.48	0.0772	1.40	0.39	2.177×10^{-2}
Trial 2	0.0384	2.0	0.0850	1.06	0.71	8.738×10^{-3}
Trial 3	0.0405	1.99	0.0826	1.02	0.73	8.120×10^{-3}
Trial 4	0.0302	1.63	0.0790	1.41	0.45	2.278×10^{-2}

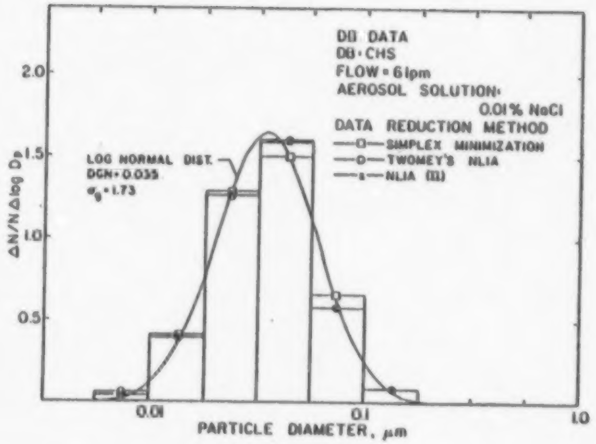

Figure 4.10(a) Histogram of the normalized number distribution of 0.01% NaCl solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 6 lpm

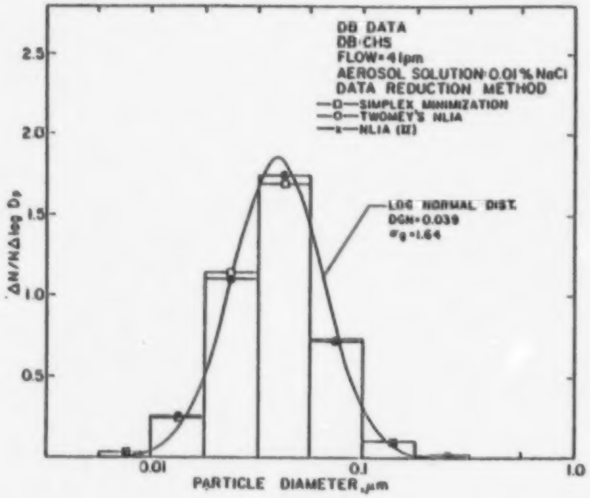

Figure 4.10(b) Histogram of the normalized number distribution of 0.01% NaCl solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 4 lpm

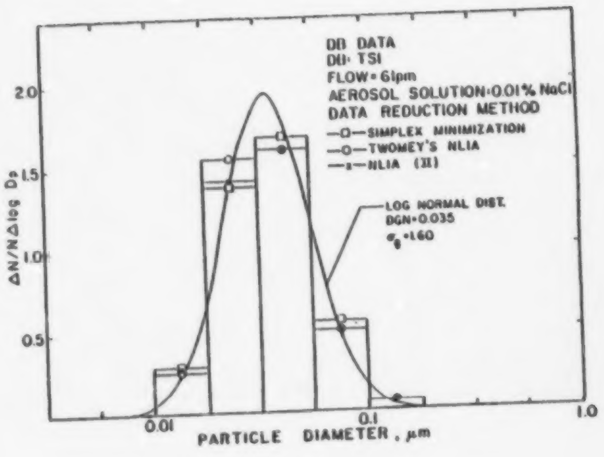

Figure 4.10(c) Histogram of the normalized number distribution of 0.01% NaCl solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 6 lpm

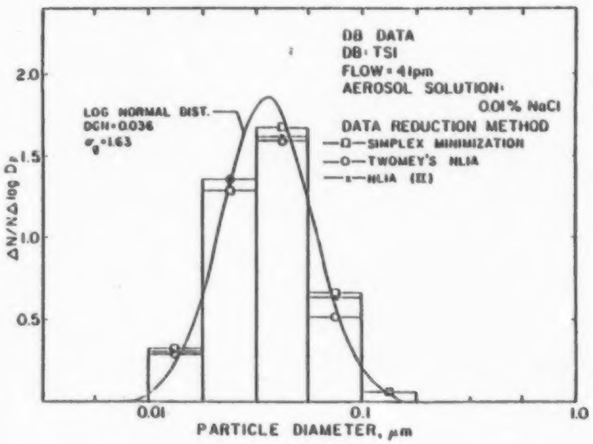

Figure 4.10(d) Histogram of the normalized number distribution of 0.01% NaCl solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 4 lpm

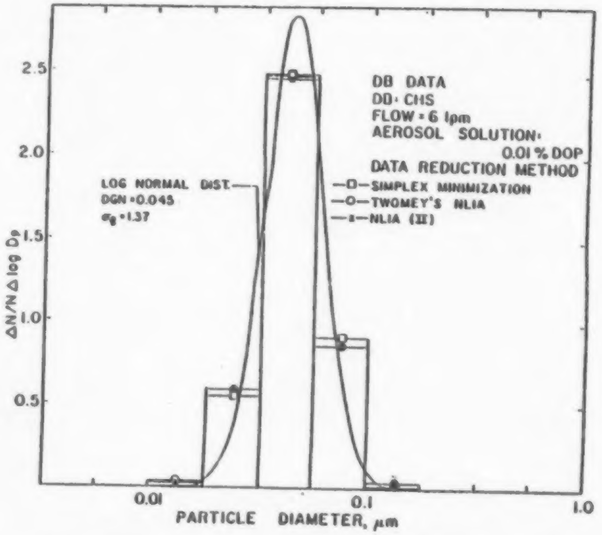

Figure 4.11(a) Histogram of the normalized number distribution of 0.01% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 6 lpm

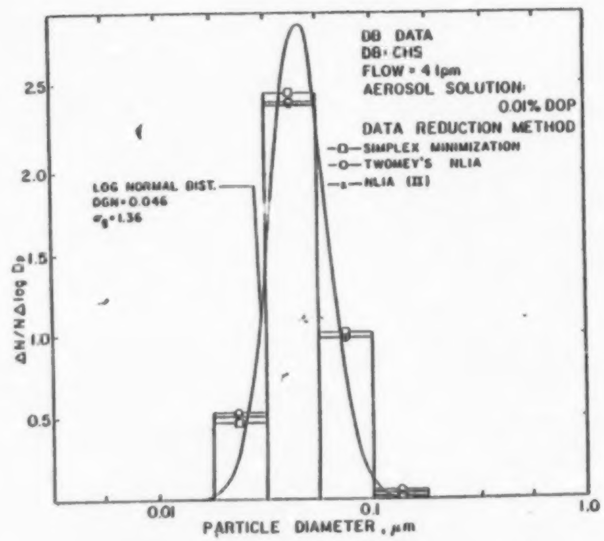

Figure 4.11(b) Histogram of the normalized number distribution of 0.01% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 4 lpm

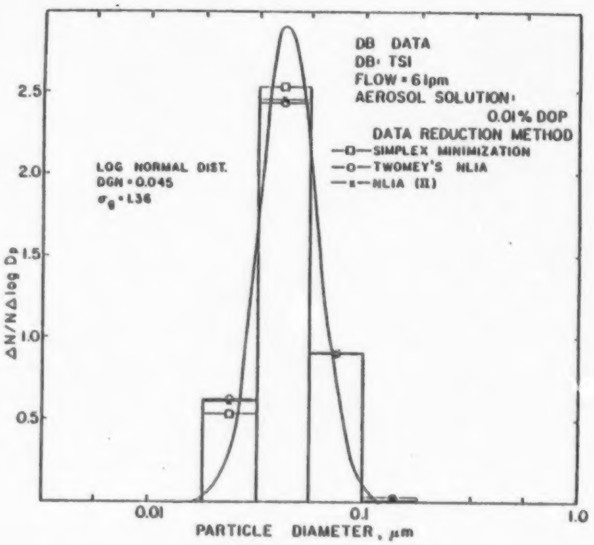

Figure 4.11(c) Histogram of the normalized number distribution of 0.01% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 6 lpm

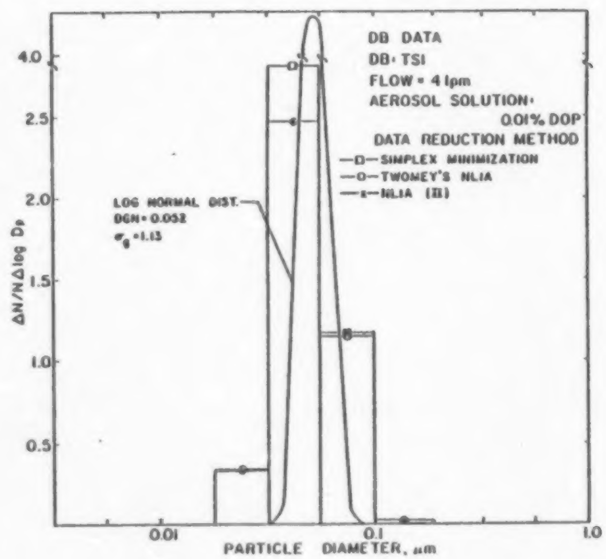

Figure 4.11(d) Histogram of the normalized number distribution of 0.01% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 4 lpm

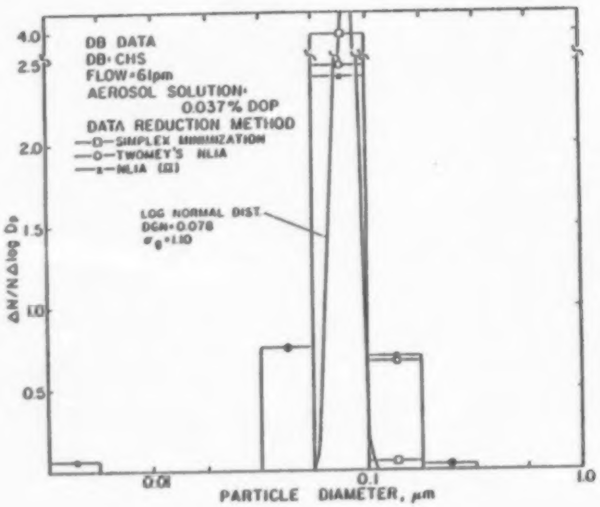

Figure 4.12(a) Histogram of the normalized number distribution of 0.037% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 6 lpm

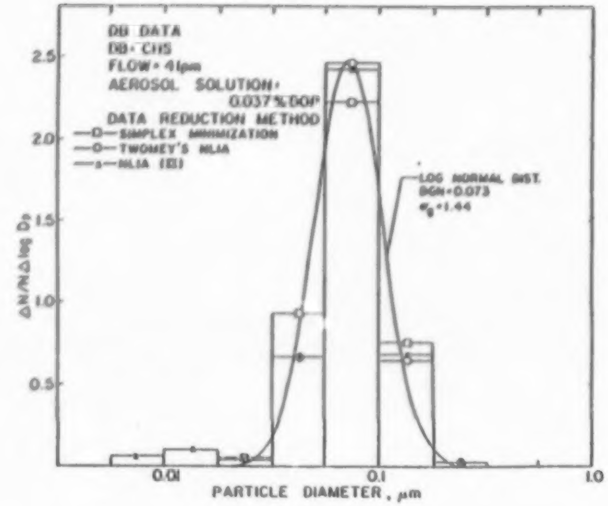

Figure 4.12(b) Histogram of the normalized number distribution of 0.037% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 4 lpm

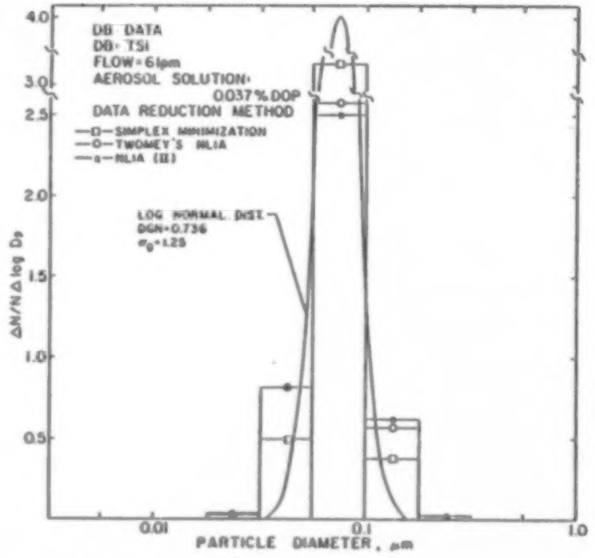

Figure 4.12(c) Histogram of the normalized number distribution of 0.037% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 6 lpm

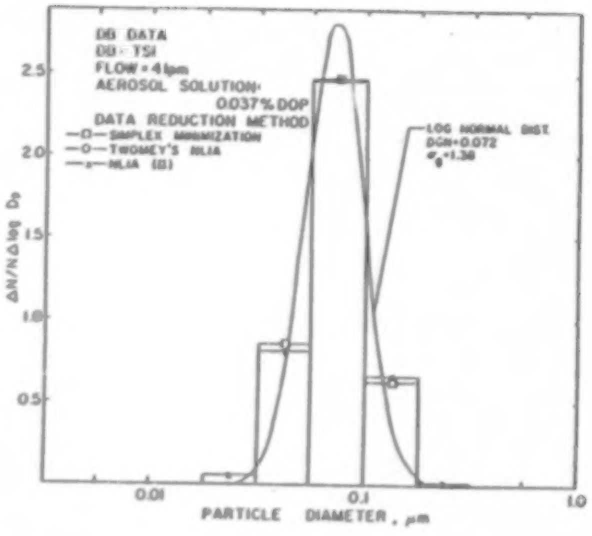

Figure 4.12(d) Histogram of the normalized number distribution of 0.037% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 4 lpm

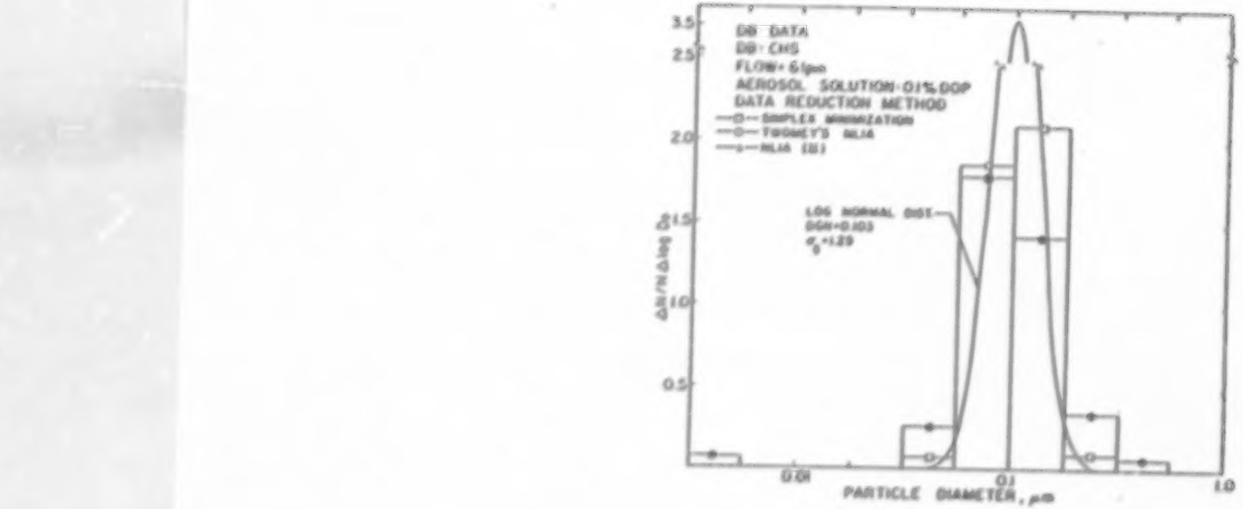

Figure 4.13(a) Histogram of the normalized number distribution of 0.1% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 6 lpm

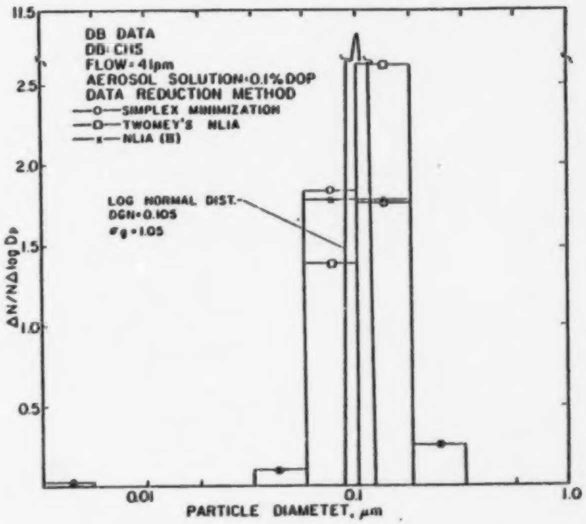

Figure 4.13(b) Histogram of the normalized number distribution of 0.1% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: CHS battery, flow: 4 lpm

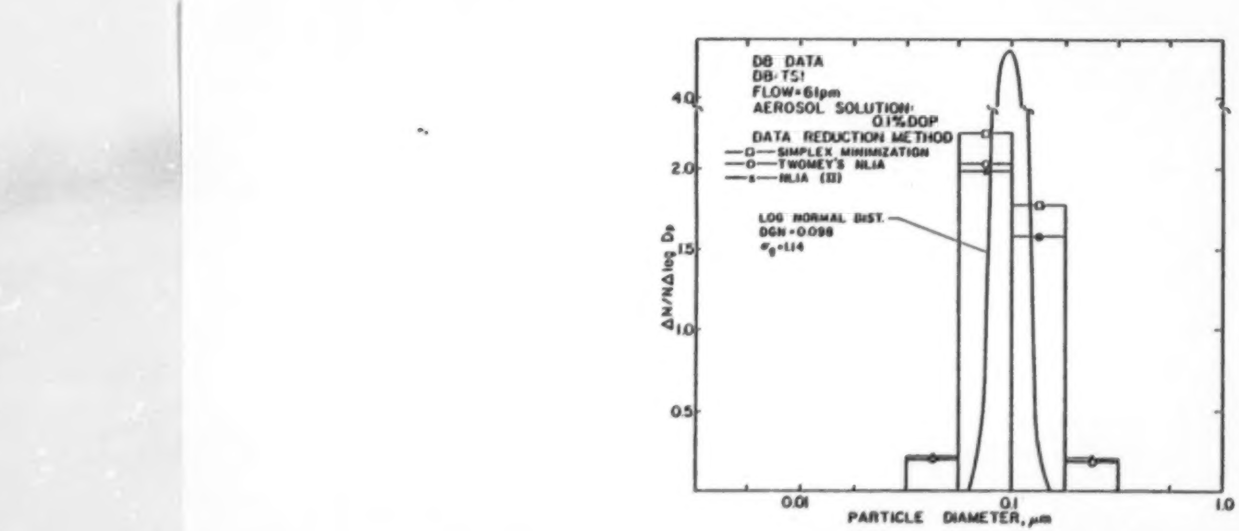

Figure 4.13(c) Histogram of the normalized number distribution of 0.1% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 6 lpm

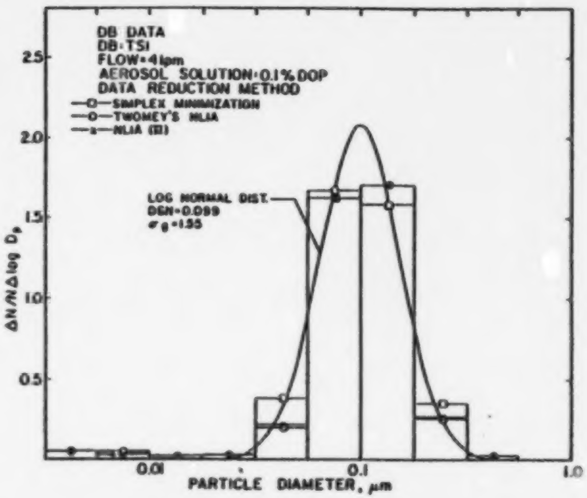

Figure 4.13(d) Histogram of the normalized number distribution of 0.1% DOP solution aerosol measured with the diffusion batteries and obtained by different data reduction techniques: TSI battery, flow: 4 lpm

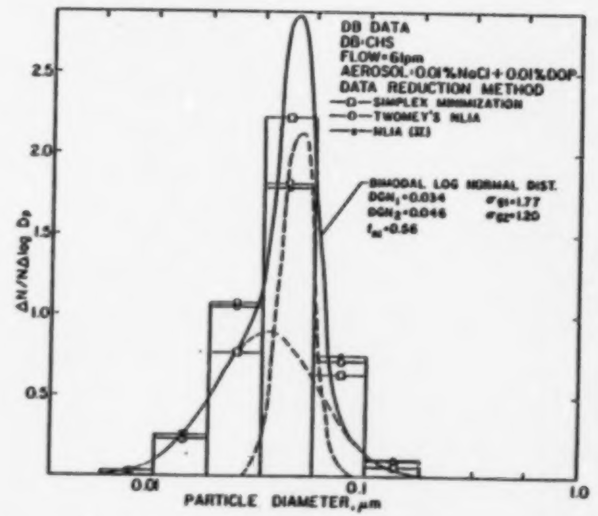

Figure 4.14 Histogram of the normalized composite number distribution of 0.01% NaCl + 0.01% DOP solution aerosol measured with CHS battery (flow: 6 lpm) and obtained by different data reduction techniques.

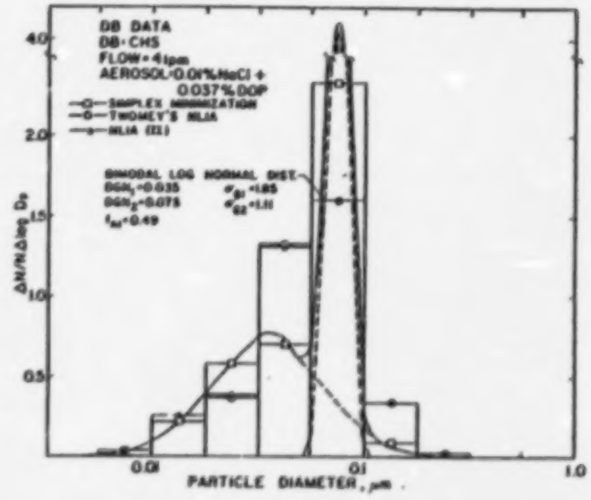

Figure 4.15 Histogram of the normalized composite number distribution of 0.01% NaCl + 0.037% DOP solution aerosol measured with CHS battery (flow: 4 lpm) and obtained by different data reduction techniques.

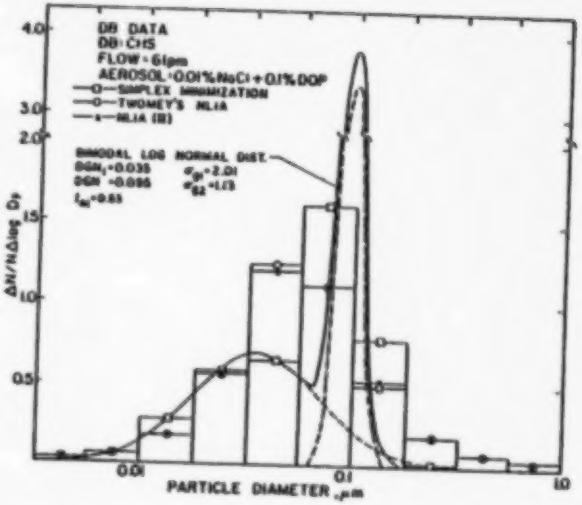

Figure 4.16 Histogram of the normalized composite number distribution of 0.01% NaCl + 0:1% DOP solution aerosol measured with CNS battery (flow: 6 lpm) and obtained by different data reduction techniques.

4.4.1 Comparison of EAA Data Reduction Methods

Figures 4.2(a), 4.3(a), 4.4(a), and 4.5(a) show that the agreement between the normalized number distribution obtained by Twomey's NLIA, NLIA(II), and the simplex minimization method is good. However, there is a difference between the distribution obtained by using the monodisperse sensitivity method of Liu and Pui (1975) and the other methods. Similarly, from Table 4.1 it can be seen that the total concentration estimates obtained by the monodisperse sensitivity method are, on the average, 7% higher than those obtained by the other three methods.

Figures 4.2(b), 4.3(b), 4.4(b), and 4.5(b) show the normalized retrieved surface distribution for the four aerosols. Once again, the surface distribution obtained by the monodisperse sensitivity method is different from the ones obtained by the other three methods. Moreover, for both non-linear iterative algorithms and the monodisperse sensitivity method, there is a significant fraction of the surface distribution in the last channel, with the fraction increasing as the mean diameter increases. From Figure 2.10, it can be seen that even for the simulation runs, the NLIA's tend to overestimate the surface concentration in the last two channels of the EAA. Also, Clark and Avol (1979) have reported that the comparison between the different EAA's for the last two channels is not as good as for the other channels of the EAA. Thus, there is considerable error in the measurement of current in the last two channels, and so the performance of the EAA, as modeled by the 11 x 45 matrix of Liu et al. (1979) may not be accurate for all the EAA's manufactured by TSI, Inc. Furthermore, the diminishing slope of the electrical mobility vs. particle size curve for large particles places a limit on the resolution capabilities of the EAA for those sizes. It can then be surmised that the anomalies noted in the surface distribution retrieved by the NLIA's can be attributed both to the data reduction procedure and the performance of the EAA for particles in the size range of the last two channels.

However, for the simplex minimization technique, these anomalies are not present because of the log-normal assumption of the form of the aerosol size distribution. The total surface concentration estimates (Table 4.1) for the simplex minimization method are also consistently lower than those obtained by the other data reduction methods, for all the four aerosols.

Similar observations, though more pronounced, can also be noted from the normalized volume distributions shown in Figures 4.2(c), 4.3(c), 4.4(c), and 4.5(c), and the estimates of the total concentration presented in Table 4.1. Also, Mulholland et al. (1979) have found that the total mass, based on the total volume obtained by the simplex minimization method for EAA data, compares satisfactorily with the gravimetric method for DOP aerosols.

For the bimodal aerosols, the results obtained by the monodisperse sensitivity method differ from the distributions obtained by the other three methods; however, the comparison between the non-linear iterative methods and the simplex minimization method is satisfactory. Observations similar to that of unimodal data can be made for the surface and volume distributions.

On examining Table 4.3, it can be noted that the parameters for the bimodal data compare favorably for those obtained with the unimodal data, except for the second bimodal aerosol for the EAA. For this aerosol, using different starting estimates for the curve fitting procedure, different values for the final parameters were obtained, even though the final values of WLS for all the trials are comparable. However, the histograms for all the solutions compare favorably, as can be seen from Figure 4.9.

4.4.2 Comparison of Diffusion Battery-CNC Data Reduction Methods

Figures 4.10 to 4.16 show good agreement between the number distributions retrived by Twomey's NLIA, NLIA(II), and the simplex minimization method for all the aerosols. From the last four columns of Table 4.2, it can be seen that the geometric number mean diameters by the simplex minimization method agree satisfactorily. However, the agreement in the various estimates of the geometric standard deviation becomes unsatisfactory as the DGN increases. Also, the standard error estimates of σ_g for the diffusion battery-CNC technique are much larger than the standard error estimates of σ_g for the EAA. Moreover, the standard error estimates of σ_g for the diffusion battery method also increase as the geometric number mean diameters increase. This increase in the uncertainty of σ_g is due to the performance of the diffusion battery, as can be seen from Figures 4.17(a) and (b). In these figures, the open symbols represent the simulated percentage loss in each stage of the diffusion battery for a given DGN but different geometric standard deviations, and the closed symbols represent the measured percentage loss in each stage

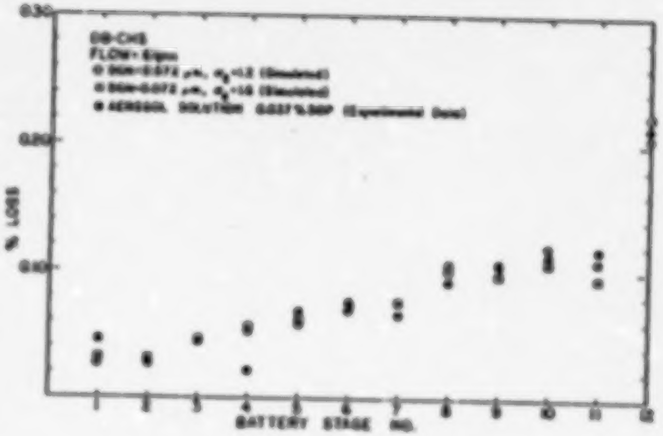

Figure 4.17(a) Comparison of simulated and measured responses of diffusion battery + CNC: CHS battery (flow: 6 lpm)

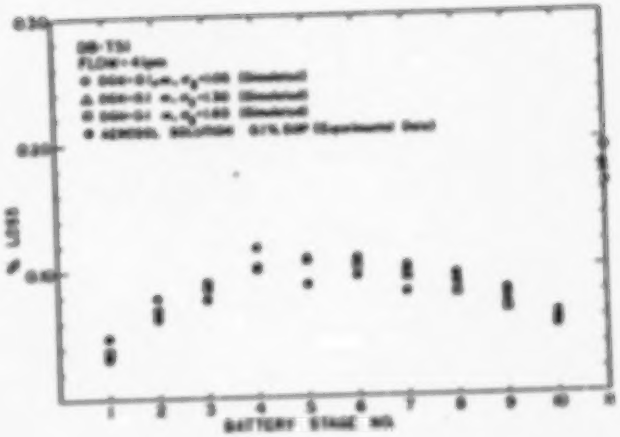

Figure 4.17(b) Comparison of simulated and measured responses of diffusion battery + CNC: TSI battery (flow: 4 lpm)

of the diffusion battery for 0.037% DBP and 0.1% DBP solution aerosols, respectively. The open symbols in both the figures show that the response of the diffusion battery does not change significantly, though there is a considerable spread in the values of σ_g. Thus, the resolution of σ_g with the diffusion battery method is poor. Similar observations were made for all the other data measured with the diffusion batteries. For the geometric number mean diameters, however, the diffusion battery method has a fair degree of resolution.

4.4.3 Comparison of EAA and the Diffusion Battery-CNC Technique

On examining Table 4.2, it can be seen that the geometric number mean diameters estimated by the simplex minimization method for both aerosol size distribution measuring techniques agree satisfactorily, while the agreement between the geometric standard deviations becomes worse as the DGN increases. As all the DBP aerosols were produced by the same generator with the vaporizer/condenser tube in line, it is reasonable to assume that all the DBP aerosols have nearly equal geometric standard deviations but different geometric number mean diameters. The estimates of σ_g from EAA measurements for DBP aerosols agree satisfactorily, but the estimates from the diffusion battery measurements do not compare favorably, especially for the larger geometric number mean diameter aerosols.

The normalized number distributions determined by the EAA and the diffusion battery on the overall compare satisfactorily for the 0.01% NaCl and 0.01% DBP aerosol, while for the 0.037% DBP and 0.1% DBP, the agreement can be considered only fair.

CHAPTER 5

SUMMARY AND CONCLUSIONS

The diffusion battery (DB) with the condensation nuclei counter (CNC) and the electrical aerosol analyzer (EAA) have been used for measuring the aerosol size distribution for some time now, but no systematic comparison of their measurement capabilities has been carried out to date, due to lack of a generalized data reduction procedure which could be applied to such instruments.

As presented earlier, the EAA and DB+CNC can be represented by Fredholm Integral Equations of the First Kind, which can be discretized to give a set of linear equations. These sets of equations are ill-conditioned, i.e., an infinitesimal change in the data vector or the coefficient matrix produces finite changes in the solution to the set of equations. Thus, if a simple inversion procedure is applied to the data measured by such instruments, a highly oscillating solution is obtained which is physically unrealistic.

Twomey (1975) developed a non-linear iterative algorithm (NLIA) for solving such a set of equations. During the course of this thesis, another non-linear iterative algorithm was developed, which was faster than Twomey's NLIA. Both these NLIA's have been adopted for solving the DB+CNC and EAA data reduction problems.

Another method developed for solving the set of linear equations consists of fitting either a unimodal or a bimodal log-normal aerosol size distribution to the measured data using Nelder and Mead's (1965) simplex minimization method. This method has also been adapted to solve the DB and EAA data reduction problem.

For the EAA, an optimized channel method was also developed, but it can be used only for specialized applications of the EAA, specifically only when the particle diameter is between 0.0075 μm to 0.19 μm for the aerosol.

These data reduction procedures were first tested in two sets of simulation studies. For the first set, the data reduction procedures for the EAA were tested with several simulated unimodal and bimodal log-normal distributions, and the DB procedures were tested with unimodal log-normal distributions. For the second set, normal random noise (mean 0, and a standard deviation of 5%, 10%, and 15%) to simulate the measurement error and the instability of the aerosol being measured was added to the expected response of the instruments for the simulated aerosol. This modified response was then used as input to the EAA and DB data reduction procedures. The retrieved distributions for the second set were compared with results obtained from the first set of simulations and were found to be within ±10% (corresponding to ±2 limits).

Finally, submicron aerosols of di-octyl phthalate (DOP) and NaCl, generated using a Collison atomizer, were simultaneously measured by the EAA and DB+CNC. The data thus obtained were analyzed using the data reduction methods developed during the course of this thesis.

For the EAA, the agreement between the various data reduction techniques for the number distribution was good. For the surface and volume distributions, however, the NLIA's and the monodisperse sensitivity method of Liu and Pui (1975) consistently overestimate the surface and volume concentration in the last two channels. The simplex minimization curve-fitting procedure, however, gives satisfactory results for all the three weightings.

The agreement between the distributions retrieved by the different data reductions for the diffusion battery was also found to be good. From the analysis of the simplex minimization method results, it can be noted that the diffusion battery is capable of resolving the geometric number mean diameter of the aerosol with a small uncertainty compared to its resolution capabilities for the geometric standard deviation. Moreover, as the geometric number mean diameter of an aerosol increases, the standard error estimates of σ_g for the diffusion battery also increase.

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APPENDIX A

RESPONSE OF THE EAA FOR MONODISPERSE AEROSOLS
[ORIGINAL DATA OF FIGURE 2.7 (LIU AND PUI, 1975)]

Diameter (µm)	0.018*		0.024		0.032	
	Volts	Percent Current	Volts	Percent Current	Volts	Percent Current
	100	100.0	60	100.0	100	100.0
	400	100.0	200	100.0	800	100.0
	500	92.9	500	97.1	900	96.8
	530	85.7	600	91.4	1000	85.8
	550	71.4	700	91.4	1100	76.3
	570	64.3	800	90.0	1200	72.6
	588	50.0	900	87.1	1300	70.5
	600	50.0	1000	71.4	1400	70.5
	630	28.6	1040	60.0	1500	70.5
	650	14.3	1062	50.0	1600	70.0
	680	3.6	1100	38.6	1700	66.8
			1150	25.7	1800	58.9
			1200	15.7	1900	48.9
			1250	8.6	2000	34.2
			1300	2.9	2200	13.7
					2400	4.3
					2600	1.6

* For diameters smaller than 0.018 µm, the slope of the current vs. voltage curve is equal to that of 0.018 µm, and the procedure for determining the voltage corresponding to [illegible]% current is given in Liu and Pui (1975).

Response of the EAA for monodisperse aerosols
[original data of Figure 2.7 (Liu and Pui, 1975)]
(continued)

Diameter (µm)	0.042		0.056		0.075	
	Volts	Percent Current	Volts	Percent Current	Volts	Percent Current
	100	100.0	200	100.0	1000	100.0
	800	100.0	1200	100.0	1700	100.0
	1000	98.6	1400	99.5	2000	98.5
	1100	97.6	1600	95.2	2300	93.6
	1200	95.9	1800	87.6	2600	85.8
	1300	93.2	2000	82.6	3000	65.5
	1400	85.6	2250	77.4	3300	53.5
	1500	73.6	2500	58.1	3600	48.7
	1600	63.0	2600	50.0	3500	50.0
	1700	55.8	2800	35.5	4000	39.4
	1800	53.1	3000	28.3	4200	32.1
	2000	50.0	3200	24.8	4500	21.0
	2200	49.3	3500	21.4	4800	13.5
	2400	47.9	3700	20.7	5000	10.4
	2600	44.5	4000	19.5	5300	7.7
	2800	34.6	4200	18.8	5600	6.2
	3000	22.9	4500	17.1	6000	5.3
	3200	11.3	4700	14.8	6300	4.4
	3500	3.8	5000	10.0	6600	4.0
	3700	1.7	5200	7.6	7000	3.8
	4000	1.0	5500	4.5	7500	3.3
			5700	2.4	8000	2.4
			6000	1.0	8500	1.8
			6300	0.4	9000	0.7
					9500	0.2

Response of the EAA for monodisperse aerosols
[original data of Figure 2.7 (Liu and Pui, 1975)]
(continued)

Diameter (µm)	0.1		0.13		0.18	
	Volts	Percent Current	Volts	Percent Current	Volts	Percent Current
	300	100.0	1000	100.0	2000	100.0
	2000	100.0	3000	100.0	4000	100.0
	3000	94.7	3500	93.4	5000	91.9
	3500	80.1	4000	79.4	5500	84.7
	4000	62.6	4500	57.9	6000	68.5
	4440	50.0	4770	50.0	6500	52.3
	5000	29.5	5000	40.8	7000	37.8
	5500	17.2	5500	25.4	7500	25.6
	6000	12.6	6000	14.9	8000	18.0
	6500	9.3	6500	9.6	8500	11.0
	7000	5.6	7000	6.1	9000	6.8
	7500	3.0	7500	3.5	10000	2.9
	8000	1.3	8000	1.8	11000	0.9
			8500	1.0		
			9000	0.6		

Response of the EAA for monodisperse aerosols
[original data of Figure 2.7 (Liu and Pui, 1975)]
(continued)

Diameter (µm)	0.24		0.32		0.42	
	Volts	Percent Current	Volts	Percent Current	Volts	Percent Current
	1000	100.0	1000	100.0	3000	100.0
	5000	100.0	5000	100.0	6500	100.0
	6000	89.4	6000	98.1	7000	96.1
	6500	81.4	6500	94.4	7500	92.1
	7000	67.4	7000	86.0	8000	81.6
	7500	53.0	7500	73.8	8500	65.8
	7600	50.0	8000	58.8	9140	50.0
	8000	37.9	8380	50.5	9500	35.5
	8500	27.7	9000	34.6	10000	23.7
	9000	17.8	9500	22.4	11000	9.2
	9500	11.7	10000	14.0		
	10000	7.6	11000	4.7		
	11000	3.0				

Response of the EAA for monodisperse aerosols
[original data of Figure 2.7 (Liu and Pui, 1975)]
(continued)

Diameter (µm)	0.56		0.75	
	Volts	Percent Current	Volts	Percent Current
	1000	100.0	5000	100.0
	5000	100.0	8000	92.5
	7000	98.3	8500	87.5
	7500	95.4	9000	77.0
	8000	88.2	9500	62.0
	8500	79.7	10000	53.0
	9000	65.4	11000	36.0
	9640	50.2	11500	18.0
	10000	39.2		
	11000	16.0		

APPENDIX B

COMPUTER PROGRAM FOR EAA DATA REDUCTION USING TWOMEY'S NON-LINEAR ITERATIVE ALGORITHM

(EAATIA)

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:44:17            PAGE 001
CORE=08K, UIC=[100,103]                                      ,LP:=EAATIA.ALL

      C * PROGRAM EAATIA.FTN
      C
      C EAATIA USES TWOMEY'S NONLINEAR ITERATIVE
      C ALGORITHM (J. OF COMPUT. PHYS. 18:188-200)
      C TO FIT A PARTICLE SIZE HISTOGRAM TO EAA DATA
      C USING THE 11X45 MATRIX. THE PROGRAM STOPS THE
      C ITERATIVE PROCEDURE EITHER WHEN THE WLS
      C IS LESS THAN .003 OR WHEN THE NUMBER
      C OF ITERATIONS BECOMES 100, WHICHEVER
      C COMES FIRST.
      C
      C DISCRIPTION OF KEY VARIBLES
      C
      C   PHI(I,J)     CALIBERATION MATRIX ELEMENT FOR
      C                THE ITH CHANNEL AND THE JTH
      C                PARTICLE SIZE
      C   SENS(J)      SENSITIVITY OF THE EAA(PA/PART/CC)
      C                FOR THE JTH SIZE
      C   DIA(J)       DIAMETER (MICRONS) OF THE JTH PARTICLE
      C                SIZE
      C   CUR1(I)      THE MEASURED CURRENT IN THE ITH CHANNEL
      C   CUR2(I)      THE CALCULATED CURRENT IN THE ITH CHANNEL
      C
      C   PC1(J) AND PC2(J) ARE INTERMEDIATE VARIABLES USED IN THE
      C   PROGRAM
      C
0001          COMMON /EAATWO/PHI(11,45)
0002          COMMON DIA(45),SENS(45),CARD(10),CUR1(11),CUR2(11),PC1(45),
             1 PC2(45)
      C
      C INITIALISE AND CALCULATE THE DIA. AND SENS.
      C
0003          CALL ASSIGN(6,'LP:')
0004          JMAX=45
0005          IMAX=11
0006          DIA(1)=.00365
0007          DPMULT=1.154781985
0008          DO 11 J=2,JMAX
0009    11    DIA(J)=DIA(J-1)*DPMULT
0010          SPACE=ALOG10(DIA(2)/DIA(1))
0011          DO 140 J=1,JMAX
0012          IF(DIA(J)-.0125)120,130,130
0013    120   SENS(J)= 2.351E6*(DIA(J))**6.262
0014          GO TO 140
0015    130   SENS(J)=0.0004264*(DIA(J))**1.159
0016    140   CONTINUE
      C
      C READ THE INPUT DATA
      C
0017    10    WRITE(5,700)
0018    700   FORMAT(10X,'ENTER DATA DECRIPTION(40 CHARACTERS)',/)
0019          READ(5,702) CARD
0020    702   FORMAT (10A4)
0021          WRITE(5,704)
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:44:17          PAGE 002
CORE=08K, UIC=[100,103]                                    ,LP:=EAATIA.ALL

0022  704   FORMAT(10X,'ENTER 11 CURRENTS FROM EAA',/)
0023        READ(5,706) CUR1
0024  706   FORMAT(11F10.0)
0025        DO 20 J=1,JMAX
0026  20    PC2(J)=CUR1(1)/JMAX
0027        IMAX1=IMAX-1
0028        DO 30 I=1,IMAX1
0029        CUR1(I)=CUR1(I)-CUR1(I+1)
0030  30    CUR1(I)=AMAX1(CUR1(I),0.0)
      C
      C ITERATE
      C
0031        ITER=0
0032        KMAX=20
0033        TCUR=0.0
0034        DO 32 I=1,IMAX
0035  32    TCUR=TCUR+CUR1(I)
0036  35    DO 60 K=1,KMAX
0037        DO 40 J=1,JMAX
0038  40    PC1(J)=PC2(J)
0039        DO 60 I=1,IMAX
0040        A=0.0
0041        DO 50 J=1,JMAX
0042  50    A=A+PHI(I,J)*PC2(J)
0043        IF(A) 60,60,54
0044  54    A=CUR1(I)/A-1.0
0045        DO 61 J=1,JMAX
0046  61    PC2(J)=PC2(J)*(1.0+A*PHI(I,J))
0047  60    CONTINUE
0048        DO 70 I=1,IMAX
0049        CUR2(I)=0.0
0050        DO 70 J=1,JMAX
0051  70    CUR2(I)=CUR2(I)+PHI(I,J)*PC2(J)
0052        IF (ITER .GE. 100) GO TO 73
0054        ITER=ITER+KMAX
0055        KMAX=5
0056        CALL OBJCT(X)
0057        IF( X .LT. .005) GO TO 73
0059        GO TO 35
      C
      C CALCULATE THE VARIABLES FOR OUTPUT
      C
0060  73    DO 80 J=1,JMAX
0061  80    PC1(J)=PC2(J)/SPACE/SENS(J)
0062        WRITE(6,710) CARD,ITER
0063  710   FORMAT(///,9X,10A4//10X,'NO. OF ITERATIONS= ',I3)
0064        PC1(2)=(PC1(1)+PC1(2)+PC1(3)+0.5*PC1(4))/3.5
0065        IF(PC1(2) .LT. 1E-05) PC1(2)=0.0
0067        DO 111 J=6,38,4
0068        PC1(J)=(PC1(J-2)/2. +PC1(J-1) +PC1(J)
           1 +PC1(J+1) +  PC1(J+2)/2.0)/4.0
0069  111   IF(PC1(J) .LT. 1E-05) PC1(J)=0.0
0071        PC1(42)=(0.5*PC1(40)+PC1(41)+PC1(42)+PC1(43)
           1 +PC1(44)+PC1(45))/5.5
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:44:17          PAGE 003
CORE=08K, UIC=[100,103]                                       ,LP:=EAATIA.ALL

0072          IF(PC1(42) .LT. 1E-05) PC1(42)=0.0
0074          DO 1070 J=2,38,4
0075          PC1(J+1)=PC1(J)*3.14*(DIA(J)**2)
0076    1070  PC1(J+2)=PC1(J)*0.523*(DIA(J)**3)
0077          WRITE(6,712)
0078    712   FORMAT(//3X,'CHANNEL',4X,'DIA.',3X,
             1 'OBS. CURR.',2X,'FIT. CURR.',2X,'DNDLGD',
             1 6X,'DSDLGD',6X,'DVDLGD'/)
0079          TNUM=0.0
0080          TCUR1=0.0
0081          TCUR2=0.0
0082          TSURF=0.0
0083          TVOL=0.0
0084          I=0
0085          DO 1090 J=2,38,4
0086          I=I+1
0087          TCUR1=TCUR1+CUR1(I)
0088          TCUR2=TCUR2+CUR2(I)
0089          TNUM=TNUM+PC1(J)
0090          TSURF=TSURF+PC1(J+1)
0091          TVOL=TVOL+PC1(J+2)
0092          WRITE(6,714) I,DIA(J),CUR1(I),CUR2(I),PC1(J),PC1(J+1),
             1 PC1(J+2)
0093    714   FORMAT(7X,I2,6X,F7.5,4X,F6.3,4X,F6.3,3(4X,1PE9.3))
0094    1090  CONTINUE
0095          TNUM=TNUM/4.0
0096          TSURF=TSURF/4.0
0097          TVOL=TVOL/4.0
0098          I=I+1
0099          TCUR1=TCUR1+CUR1(I)
0100          TCUR2=TCUR2+CUR2(I)
0101          WRITE(6,716) I,CUR1(I),CUR2(I)
0102    716   FORMAT(7X,I2,17X,F6.3,4X,F6.3)
0103          WRITE(6,718) TCUR1,TCUR2,TNUM,TSURF,TVOL
0104    718   FORMAT(/3X,'TOTALS',15X,F6.3,5X,F5.3,3(4X,1PE9.3))
        D     DO 711 J=2,38,4
        D     PC1(J+1)=PC1(J+1)/(TSURF*4.)
        D     PC1(J+2)=PC1(J+2)/(TVOL*4.)
        D711  PC1(J)=PC1(J)/(TNUM*4.0)
        D     WRITE(6,717)(PC1(J),J=2,38,4)
        D     WRITE(6,717)(PC1(J+1),J=2,38,4)
        D     WRITE(6,717)(PC1(J+2),J=2,38,4)
0105    717   FORMAT(//3X,10(4X,F6.4)/)
0106          WRITE(5,720)
0107    720   FORMAT(/10X,'ENTER 1 FOR ANOTHER CALC.  2 FOR STOP',/)
0108          READ(5,722) IFLAG
0109    722   FORMAT(I5)
0110          GO TO (10,18) IFLAG
0111    18    STOP
0112          END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:45:07          PAGE 001
CORE=08K, UIC=[100,103]                                      ,LP:=EAATIA.ALL

0001        BLOCK DATA
0002        COMMON/EAATWO/PHI(11,45)
0003        DATA PHI/
           1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
           1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
           1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .500,.500,.000,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.500,.500,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,.480,.520,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,.050,.950,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,.080,.920,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,
           1 .000,.000,.000,.510,.480,.010,.000,.000,.000,.000,.000,
           1 .000,.000,.000,.440,.520,.040,.000,.000,.000,.000,.000,
           1 .000,.000,.000,.490,.070,.440,.000,.000,.000,.000,.000,
           1 .000,.000,.000,.150,.480,.195,.175,.000,.000,.000,.000,
           1 .000,.000,.000,.145,.355,.325,.175,.000,.000,.000,.000,
           1 .000,.000,.000,.035,.345,.515,.085,.020,.000,.000,.000,
           1 .000,.000,.000,.000,.140,.650,.170,.020,.020,.000,.000,
           1 .000,.000,.000,.000,.050,.610,.310,.030,.000,.000,.000,
           1 .000,.000,.000,.000,.000,.510,.405,.085,.000,.000,.000,
           1 .000,.000,.000,.000,.000,.450,.480,.070,.000,.000,.000,
           1 .000,.000,.000,.000,.000,.400,.510,.090,.000,.000,.000,
           1 .000,.000,.000,.000,.000,.130,.670,.170,.030,.000,.000,
           1 .000,.000,.000,.000,.000,.030,.470,.380,.080,.040,.000,
           1 .000,.000,.000,.000,.000,.000,.350,.460,.140,.040,.010,
           1 .000,.000,.000,.000,.000,.000,.220,.480,.200,.060,.040,
           1 .000,.000,.000,.000,.000,.000,.150,.450,.250,.090,.060,
           1 .000,.000,.000,.000,.000,.000,.080,.420,.320,.110,.070,
           1 .000,.000,.000,.000,.000,.000,.020,.320,.380,.180,.100,
           1 .000,.000,.000,.000,.000,.000,.000,.310,.360,.180,.150,
           1 .000,.000,.000,.000,.000,.000,.000,.200,.390,.210,.200,
           1 .000,.000,.000,.000,.000,.000,.000,.195,.305,.260,.240,
           1 .000,.000,.000,.000,.000,.000,.000,.130,.270,.250,.350,
           1 .000,.000,.000,.000,.000,.000,.000,.110,.280,.250,.360,
           1 .000,.000,.000,.000,.000,.000,.000,.070,.230,.250,.450,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500/
0004        END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:45:23          PAGE 001
CORE=08K, UIC=[100,103]                                   .LP:=EAATIA.ALL

      C       ***ULS.FTN***
0001          SUBROUTINE OBJCT(X)
0002          COMMON DIA(45),SENS(45),CAPD(10),CUR1(11),CUR2(11),PC1(45),
             1 PC2(45),CHANGE(45)
0003          TCUR1=0.0
0004          TCUR2=0.0
0005          IMAX=11
0006          X=0.0
0007          DO 10I=1,IMAX
0008          TCUR1=TCUR1+CUR1(I)
0009    10    TCUR2=TCUR2+CUR2(I)
0010          DO 20 I=1,IMAX
0011          ACUR2=CUR2(I)/TCUR2
0012          AF=(CUR1(I)/TCUR1)-ACUR2
0013          IF(ABS(AF) .LT. .001) GO TO 20
0015          X=X+(AF**2)/(CUR1(I)/TCUR1+1.0E-15)
0016    20    CONTINUE
0017          RETURN
0018          END
```

APPENDIX C

COMPUTER PROGRAM FOR EAA DATA REDUCTION USING NON-LINEAR ITERATIVE ALGORITHM (II)

(EAAKIA)

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:46:18          PAGE 001
CORE=08K, UIC=[100,103]                                     .LP:=EAAKIA.ALL
      C * PROGRAM EAAKIA.FTH
      C
      C EAAKIA USES A NONLINEAR ITERATIVE ALGORITHM
      C TO FIT A PARTICLE SIZE HISTOGRAM TO EAA DATA
      C USING THE 11X45 MATRIX.THE PROGRAM STOPS THE
      C ITERATIVE PROCEDURE EITHER WHEN THE WLS
      C IS LESS THAN .003 OR WHEN THE NUMBER
      C OF ITERATIONS BECOMES 100, WHICHEVER
      C COMES FIRST.
      C
      C DISCRIPTION OF KEY VARIBLES
      C
      C   PHI(I,J)     CALIBERATION MATRIX ELEMENT FOR
      C                THE ITH CHANNEL AND THE JTH
      C                PARTICLE SIZE
      C   SENS(J)      SENSITIVITY OF THE EAA(PA/PART/CC)
      C                FOR THE JTH SIZE
      C   DIA(J)       DIAMETER (MICRONS) OF THE JTH PARTICLE
      C                SIZE
      C   CUR1(I)      THE MEASURED CURRENT IN THE ITH CHANNEL
      C   CUR2(I)      THE CALCULATED CURRENT IN THE ITH CHANNEL
      C
      C   PC1(J) AND PC2(J) ARE INTERMEDIATE VARIABLES USED IN THE
      C   PROGRAM
      C
              COMMON /EAATWO/PHI(11,45)
              COMMON DIA(45),SENS(45),CARD(10),CUR1(11),CUR2(11),PC1(45),
             1 PC2(45)
      C
      C INITIALISE AND CALCULATE THE DIA. AND SENS.
      C
              CALL ASSIGN(6,'LP:')
              JMAX=45
              IMAX=11
              DIA(1)=.00365
              DPMULT=1.154781985
              DO 11 J=2,JMAX
        11    DIA(J)=DIA(J-1)*DPMULT
              SPACE=ALOG10(DIA(2)/DIA(1))
              DO 140 J=1,JMAX
              IF(DIA(J)-.0125)120,130,130
        120   SENS(J)= 2.351E6*(DIA(J))**6.262
              GO TO 140
        130   SENS(J)=0.0004264*(DIA(J))**1.159
        140   CONTINUE
      C
      C READ THE INPUT DATA
      C
        10    WRITE(5,700)
        700   FORMAT(10X,'ENTER DATA DECRIPTION(40 CHARACTERS)',/)
              READ(5,702) CARD
        702   FORMAT (10A4)
              WRITE(5,704)
        704   FORMAT(10X,'ENTER 11 CURRENTS FROM EAA',/)
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:46:18          PAGE 002
CORE=08K, UIC=[100,103]                                     ,LP:=EAAKIA.ALL

0023            READ(5,706) CUR1
0024     706    FORMAT(11F10.0)
0025            DO 20 J=1,JMAX
0026     20     PC2(J)=CUR1(1)/JMAX
0027            IMAX1=IMAX-1
0028            DO 30 I=1,IMAX1
0029            CUR1(I)=CUR1(I)-CUR1(I+1)
0030     30     CUR1(I)=AMAX1(CUR1(I),0.0)
         C
         C ITERATE
         C
0031            ITER=0
0032            KMAX=20
0033            TCUR=0.0
0034            DO 32 I=1,IMAX
0035     32     TCUR=TCUR+CUR1(I)
0036            DO 50 I=1,IMAX
0037            CUR2(I)=0.0
0038            DO 50 J=1,JMAX
0039     50     CUR2(I)=CUR2(I)+PHI(I,J)*PC2(J)
0040     35     DO 60 K=1,KMAX
0041            DO 40 J=1,JMAX
0042            CORR=0.0
0043            DO 71 I=1,IMAX
0044            IF (CUR2(I) .LT. 1.0E-20) CUR2(I)=1.0E-20
0046            IF (CUR1(I) .LT. .0005) CUR1(I)=0.0
0048     71     CORR=CORR+(CUR1(I)/CUR2(I))*PHI(I,J)
0049     40     PC2(J)=CORR*PC2(J)
0050            DO 70 I=1,IMAX
0051            CUR2(I)=0.0
0052            DO 70 J=1,JMAX
0053     70     CUR2(I)=CUR2(I)+PHI(I,J)*PC2(J)
0054     60     CONTINUE
0055            IF (ITER .GE. 100) GO TO 73
0057            ITER=ITER+KMAX
0058            KMAX=5
0059            CALL OBJECT(X)
0060            IF( X .LT. .005) GO TO 73
0062            GO TO 35
         C
         C CALCULATE THE VARIABLES FOR OUTPUT
         C
0063     73     DO 80 J=1,JMAX
0064     80     PC1(J)=PC2(J)/SPACE/SENS(J)
0065            WRITE(6,710) CARD,ITER
0066     710    FORMAT(///,9X,10A4//10X,'NO. OF ITERATIONS= ',I3)
0067            PC1(2)=(PC1(1)+PC1(2)+PC1(3)+0.5*PC1(4))/3.5
0068            IF(PC1(2) .LT. 1E-05) PC1(2)=0.0
0070            DO 111 J=6,38,4
0071            PC1(J)=(PC1(J-2)/2. +PC1(J-1) +PC1(J)
              1 +PC1(J+1) +  PC1(J+2)/2.0)/4.0
0072     111    IF(PC1(J) .LT. 1E-05) PC1(J)=0.0
0074            PC1(42)=(0.5*PC1(40)+PC1(41)+PC1(42)+PC1(43)
              1 +PC1(44)+PC1(45))/5.5
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:46:18          PAGE 002
CORE=08K, UIC=[100,103]
                                                         .LP:=EAAKIA.ALL

0076           IF(PC1(42) .LT. 1E-05) PC1(42)=0.0
0077           DO 1070 J=2,38,4
0078           PC1(J+1)=PC1(J)*3.14*(DIA(J)**2)
0079     1070  PC1(J+2)=PC1(J)*0.523*(DIA(J)**3)
0080           WRITE(6,712)
0081     712   FORMAT(//5X,'CHANNEL',4X,'DIA.',3X,
              1 'OBS. CURR.',2X,'FIT. CURR.',2X,'DNDLGD',
              1 6X,'DSDLGD',6X,'DVDLGD'/)
0082           TNUM=0.0
0083           TCUR1=0.0
0084           TCUR2=0.0
0085           TSURF=0.0
0086           TVOL=0.0
0087           I=0
0088           DO 1090 J=2,38,4
0089           I=I+1
0090           TCUR1=TCUR1+CUR1(I)
0091           TCUR2=TCUR2+CUR2(I)
0092           TNUM=TNUM+PC1(J)
0093           TSURF=TSURF+PC1(J+1)
0094           TVOL=TVOL+PC1(J+2)
0095           WRITE(6,714) I,DIA(J),CUR1(I),CUR2(I),PC1(J),PC1(J+1),
              1 PC1(J+2)
0096     714   FORMAT(7X,I2,6X,F7.3,4X,F6.3,4X,F6.3,3(4X,1PE9.3))
0097     1090  CONTINUE
0098           TNUM=TNUM/4.0
0099           TSURF=TSURF/4.0
0100           TVOL=TVOL/4.0
0101           I=I+1
0102           TCUR1=TCUR1+CUR1(I)
0103           TCUR2=TCUR2+CUR2(I)
0104           WRITE(6,716) I,CUR1(I),CUR2(I)
0105     716   FORMAT(7X,I2,17X,F6.3,4X,F6.3)
0106           WRITE(6,718) TCUR1,TCUR2,TNUM,TSURF,TVOL
0107     718   FORMAT(/5X,'TOTALS',15X,F6.3,5X,F5.3,3(4X,1PE9.3))
         D     DO 711 J=2,38,4
         D     PC1(J+1)=PC1(J+1)/(TSURF*4.)
         D     PC1(J+2)=PC1(J+2)/(TVOL*4.)
         D711  PC1(J)=PC1(J)/(TNUM*4.0)
         D     WRITE(6,717)(PC1(J),J=2,38,4)
         D     WRITE(6,717)(PC1(J+1),J=2,38,4)
         D     WRITE(6,717)(PC1(J+2),J=2,38,4)
         D717  FORMAT(//5X,10(4X,F6.4)/)
0108           WRITE(5,720)
0109     720   FORMAT(/10X,'ENTER 1 FOR ANOTHER CALC. 2 FOR STOP',/)
0110           READ(5,722) IFLAG
0111     722   FORMAT(I3)
0112           GO TO (10,18) IFLAG
0113     18    STOP
0114           END
```

```
FORTRAN IV      V01C-03B             THU 16-NOV-79 11:47:08                  PAGE 001
CORE=08K, UIC=[200,103]                                              ,LP:=EAAKIA.ALL
0001        BLOCK DATA
0002        COMMON/EAATWO/PHI(11,48)
0003        DATA PHI/
          1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
          1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
          1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .500,.500,.000,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.500,.500,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,.480,.520,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,.050,.950,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,[illegible],[illegible],.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,[illegible],[illegible],.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,[illegible],[illegible],.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,
          1 .000,.000,.000,.910,.080,.010,.000,.000,.000,.000,.000,
          1 .000,.000,.000,.440,.520,.040,.000,.000,.000,.000,.000,
          1 .000,.000,.000,.490,.070,.440,.000,.000,.000,.000,.000,
          1 .000,.000,.000,.130,.480,.190,.170,.000,.000,.000,.000,
          1 .000,.000,.000,.140,[illegible],[illegible],.170,.000,.000,.000,.000,
          1 .000,.000,.000,.020,[illegible],.310,[illegible],.000,.000,.000,.000,
          1 .000,.000,.000,.000,.140,.690,.170,.000,.000,.000,.000,
          1 .000,.000,.000,.000,[illegible],.610,.310,[illegible],.000,.000,.000,
          1 .000,.000,.000,.000,.000,.310,[illegible],[illegible],.000,.000,.000,
          1 .000,.000,.000,.000,.000,.450,[illegible],.070,.000,.000,.000,
          1 .000,.000,.000,.000,.000,[illegible],.710,.090,.000,.000,.000,
          1 .000,.000,.000,.000,.000,.130,.670,.170,.030,.000,.000,
          1 .000,.000,.000,.000,.000,[illegible],.470,[illegible],.080,.040,.000,
          1 .000,.000,.000,.000,.000,.000,[illegible],.440,.160,.040,.010,
          1 .000,.000,.000,.000,.000,.000,[illegible],.480,[illegible],.060,.040,
          1 .000,.000,.000,.000,.000,.000,.130,[illegible],[illegible],.090,.040,
          1 .000,.000,.000,.000,.000,.000,[illegible],[illegible],[illegible],.110,.070,
          1 .000,.000,.000,.000,.000,.000,.000,[illegible],[illegible],.160,.100,
          1 .000,.000,.000,.000,.000,.000,.000,.210,[illegible],.180,.150,
          1 .000,.000,.000,.000,.000,.000,.000,[illegible],[illegible],.210,.200,
          1 .000,.000,.000,.000,.000,.000,.000,.170,[illegible],.240,.240,
          1 .000,.000,.000,.000,.000,.000,.000,.130,[illegible],.250,.250,
          1 .000,.000,.000,.000,.000,.000,.000,.110,[illegible],.250,.340,
          1 .000,.000,.000,.000,.000,.000,.000,.070,[illegible],.230,.450,
          1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
          1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
          1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
          1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
          1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500/
0004        END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:47:25          PAGE 001
CORE=08K, UIC=[100,103]                                    ,LP:=EAAA1A.ALL

0001            SUBROUTINE OBJCT(X)
0002            COMMON DIA(45),SENS(45),CARD(10),CUR1(11),CUR2(11),PC1(45),
               1 PC2(45),CHANGE(45)
0003            TCUR1=0.0
0004            TCUR2=0.0
0005            IMAX=11
0006            X=0.0
0007            DO 10 I=1,IMAX
0008            TCUR1=TCUR1+CUR1(I)
0009      10    TCUR2=TCUR2+CUR2(I)
0010            DO 20 I=1,IMAX
0011            ACUR2=CUR2(I)/TCUR2
0012            AF=(CUR1(I)/TCUR1)-ACUR2
0013            IF(ABS(AF) .LT. .001) GO TO 20
0015            X=X+(AF**2)/(CUR1(I)/TCUR1+1.0E-15)
0016      20    CONTINUE
0017            RETURN
0018            END
```

APPENDIX D

COMPUTER PROGRAM FOR EAA DATA REDUCTION USING SIMPLEX MINIMIZATION CURVE-FITTING PROCEDURE

(EAAWLS)

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:37:38         PAGE 001
CORE=06K, UIC=[100,103]                                   ,LP:=[illegible].ALL

      C       [illegible].FTN IS THE MAIN PROGRAM FOR DATA
      C       REDUCTION OF SAA USING THE SIMPLEX
      C       MINIMIZATION METHOD.
      C
      C       WRITTEN BY ABBE KAPADIA AUG. '79
      C
      C
      C
0001          COMMON [illegible]
             1[illegible]
             2[illegible]
             3[illegible]
             4[illegible]
             5[illegible]
      C
0002          DIMENSION TITTLE(20),ALPHA(3,3)
      C       DIMENSION [illegible]
0003          DOUBLE PRECISION ALPHA
0004          EXTERNAL OBJCT
0005          CALL ASSIGN (6,'LP:')
      C
0006          CALL [illegible]
0007          DO 100 J=1,3
0008    100   MASK(J)=0
0009          [illegible]
0010    190   WRITE(5,200)
0011    200   FORMAT(' 1X,'ENTER DATA DESCRIPTION (40 CHARACTERS MAX.)',/)
0012          READ(5,210) TITTLE
0013    210   FORMAT(20A2)
0014          WRITE(5,220)
0015    220   FORMAT(/1X,'1 OR 2 MODES?',/)
0016          READ(5,230) MODE
0017    230   FORMAT(I1)
0018          IF(MODE .EQ. 1) GO TO 240
0019          GO TO 250
0021    240   MASK(2)=1
0022          MASK(4)=1
0023          MASK(5)=1
0024    250   IF(MODE .EQ. 2) GO TO 260
0026          WRITE(5,270)
0027    270   FORMAT(/1X,'MIN. AND MAX. VALUES OF HMD1 AND SIGMA1',/)
0028          DO 280 J=1,2
0029    280   READ(5,290) XMIN(J),XMAX(J)
0030    290   FORMAT(2F12.3)
0031          XMIN(3)=0.01
0032          XMIN(4)=1.01
0033          XMIN(5)=0.0
0034          XMAX(3)=1.0
0035          XMAX(4)=2.0
0036          XMAX(5)=1.0
0037          GO TO 400
0038    260   WRITE(5,310)
0039    310   FORMAT(/1X,'MIN. AND MAX. VALUES OF HMD1,SIGMA1,',/
             1 1X,'HMD2,SIGMA2 AND NUM. FRAC. IN MODE ONE',/)
```

```
FORTRAN IV      V01C-03D       THU 16-AUG-79 11:37:35          PAGE 002
CORE=08K, UIC=[100,103]                                  .LP:=EAAWLS.ALL

0040            DO 320 J=1,5
0041    320     READ(5,330) XMIN(J),XMAX(J)
0042    330     FORMAT(2F13.0)
0043    600     WRITE(5,340)
0044    340     FORMAT(/5X,'ENTER 11 VALUES OF I FROM EAA',/)
0045            READ(5,350) DAT
0046    350     FORMAT(11F13.0)
0047            DO 360 J=1,10
0048            DAT(J)=DAT(J)-DAT(J+1)
0049            DAT(J)=AMAX1(DAT(J),0.0)
0050            IF(DAT(J) .LT. .001) GO TO 370
0052            IF(SENSS(J) .LT. .0001) GO TO 370
0054            DNUMM(J)=4.0*(DAT(J)/SENSS(J))*1.0E6
        D       DSURM(J)=DNUMM(J)*(DPIS(J)**2)*3.14
        D       DVOLM(J)=DNUMM(J)*(DPIS(J)**3)*.524
0055            GO TO 360
0056    370     DNUMM(J)=0.0
        D       DSURM(J)=0.0
        D       DVOLM(J)=0.0
0057    360     CONTINUE
0058    610     IF(MODE .EQ. 2) GO TO 390
0060            WRITE(5,400)
0061    400     FORMAT(/5X,'ESTIMATES OF NMD1 AND SIGMA1',/)
0062            READ(5,410) X(1),X(2)
0063    410     FORMAT(5F13.0)
0064            X(3)=0.1
0065            X(4)=2.0
0066            X(5)=1.0
0067            GO TO 420
0068    390     WRITE(5,430)
0069    430     FORMAT(/5X,'ESTIMATES OF NMD1,SIGMA1,'/
               1/5X,'NMD2,SIGMA2 AND NUM. FRACT. IN MODE ONE',/)
0070            READ(5,410)X
0071    420     TDAT=0.0
0072            DO 440 J=1,11
0073    440     TDAT=TDAT+DAT(J)
0074            DO 450 J=1,11
0075    450     DAT(J)=DAT(J)/TDAT
0076            CALL STEPIT
0077            DO 460 J=1,11
0078    460     DAT(J)=DAT(J)*TDAT
0079            TNUMM=0.0
        D       TSURM=0.0
        D       TVOLM=0.0
0080            DO 470 J=1,10
        D       TSURM=TSURM+DSURM(J)
        D       TVOLM=TVOLM+DVOLM(J)
0081    470     TNUMM=TNUMM+DNUMM(J)
0082            TNUMM=TNUMM/4.0
        D       TSURM=TSURM/4.
        D       TVOLM=TVOLM/4.
0083            CALL SENS2
0084            WRITE(5,630)
0085    630     FORMAT(5X,'ENTER 1 IF YOU WANT ERROR ANALYSIS',/
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:37:35          PAGE 003
CORE=08K, UIC=[100,103]                                  ,LP:=EAAWLS.ALL

             15X,'OR 2 IF YOU DO NOT',/)
0086          READ (5,620) IFLAG
0087          IF (IFLAG .EQ. 1) GO TO 640
0089          GO TO 650
0090   640    WRITE (5,660)
0091   660    FORMAT(5X,'ENTER S.E. PROPORTIONALITY CONSTANT',/
             15X,'(0.15 FOR LABORATORY AEROSOL OR 0.25 FOR ',/
             15X,'ATMOSPHERIC AEROSOL)',/)
0092          READ(5,610) CSIG
0093          CALL ERRMAT (ALPHA,MODE)
0094          DO 670 I=1,5
0095   670    ALPHA(I,I)=SQRT(ALPHA(I,I))*CSIG
0096   650    WRITE(6,480) TITTLE
0097   480    FORMAT(1H1,/5X,20A2)
0098          WRITE(6,490)BCHISQ,FCHISQ,NCOMP,MODE,ANMD1,ASMD1,AVMD1,SIG1
0099   490    FORMAT(//1X,'BEGIN OBJCT.  FCT.  VALUE='1PE9.3,9X,
             1'FINAL OBJCT.  FCT.  VALUE=',1PE9.3,/1X,'FCT.  COMPUTATIONS=',
             20PI4,20X,I1,'MODE(S)',/1X,'NMD1      =',0PF7.4,2X,
             3'SMD1      =',F7.4,2X,'VMD1      =',F7.4,3X,'SIGMA1=',F4.2)
0100          IF (IFLAG .EQ. 1 .AND. MODE .EQ. 1) WRITE(6,680) ALPHA(1,1),
             1ALPHA(2,2)
0102   680    FORMAT(/1X,'STANDARD ERROR ESTIMATES',/
             11X,'NMD1=',1PE7.1,3X,'SIGMA1=',1PE7.1)
0103          IF(MODE .EQ. 1) GO TO 500
0105          WRITE(6,510)ANMD2,ASMD2,AVMD2,SIG2,FNUM,FSUR,FVOL
0106   510    FORMAT(1X,'NMD2      =',F7.4,2X,'SMD2      =',F7.4,2X,
             1'VMD2      =',F7.4,3X,'SIGMA2=',F4.2,/
             21X,'MODE ONE=',F4.2,6X,'MODE ONE=',F4.2,6X,'MODE ONE=',F4.2)
0107          IF (IFLAG .EQ. 1) WRITE (6,690) ALPHA(1,1),ALPHA(2,2),
             1ALPHA(3,3),ALPHA(4,4),ALPHA(5,5)
0109   690    FORMAT(/1X,'STANDARD ERROR ESTIMATES',/
             11X,'NMD1='1PE7.1,3X,'SIGMA1=',1PE7.1,
             23X,'NMD2=',1PE7.1,3X,'SIGMA2=',1PE7.1,/
             21X,'NUMBER FRACTION IN FIRST MODE =',1PE7.1)
0110   500    WRITE(6,520)
0111   520    FORMAT(//1X,'CHAN',2X,'MIDPT',3X,'DELIPA',3X,'MDNDLGDP',
             13X,' DNDLGDP ',3X' DSDLGDP ',3X,' DVDLGDP ',/)
0112          DO 530 J=1,10
0113   530    WRITE(6,540)J,DPIS(J),DAT(J),DNUMM(J),DNUMP(J),DSURP(J)
             1,DVOLP(J)
0114   540    FORMAT(2X,I2,2X,F6.4,2X,F6.3,4X,1PE9.3,4X,1PE9.3,2(3X,1PE9.3))
0115          J=11
0116          WRITE(6,550)J,DAT(J)
0117   550    FORMAT(2X,I2,10X,F6.3)
0118          WRITE(6,560) TDAT,TNUMM,TNUM,TSUR,TVOL
0119   560    FORMAT(1X,'TOTL',9X,F6.3,4X,1PE9.3,4X,1PE9.3,3X,1PE9.3,3X,
             11PE9.3)
       D      DO 563 J=1,10
       D      DSURP(J)=DSURP(J)/(TSUR*4.)
       D      DVOLP(J)=DVOLP(J)/(TVOL*4.)
       D      DNUMM(J)=DNUMM(J)/(TNUMM*4.)
       D      DSURM(J)=DSURM(J)/(TSURM*4.)
       D      DVOLM(J)=DVOLM(J)/(TVOLM*4.)
       D563   DNUMP(J)=(DNUMP(J)/TNUM)*.25
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:37:35         PAGE 004
CORE=08K, UIC=[100,103]                                    .LP:=EAAMLS.ALL

      D      WRITE(6,565) DNUMP,DSURP,DVOLP,DNUMM,DSURM,DVOLM
      D      WRITE (6,1023) TNUMM,TSURM,TVOLM
      D565   FORMAT(//5X,10(4X,F6.4),/)
      D1023  FORMAT(4(3X,1PE9.3))
0120         WRITE(5,570)
0121  570    FORMAT(/5X,'ENTER 1 TO STOP',/5X,
            1'2 TO START FROM BEGINNING',/5X,
            3'3 TO ENTER NEW CURRENTS AND ESTIMATES',/5X,
            4'4 TO ENTER NEW ESTIMATES FOR THE SAME DATA',/)
0122         READ (5,620) IFLAG
0123  620    FORMAT(I1)
0124         GO TO (380,590,600,610) IFLAG
0125  380    STOP
0126         END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:38:36          PAGE 001
CORE=08K, UIC=[100,103]                                  ,LP:=EAAULS.ALL

      C       *SENS1.FTN CALCULATES THE MONODISPERSE SENSITIVITIES
      C       FROM THE REGRESSION EQNS.
      C
0001          SUBROUTINE SENS1
0002          COMMON SCHISQ,FCHISQ,NCOMP,FNUM,FSUR,FVOL,
             1ANMD1,ANMD2,ASMD1,ASMD2,AVMD1,AVMD2,SIG1,SIG2,
             2DNUMP(10),DSURP(10),DVOLP(10),DNUMM(10),
             3TNUM,TSUR,TVOL,CALIB(11,45),SENS(45),SENSS(10),
             4DPIS(10),SDS(11),DAT(11),TDAT,
             5MASK(5),X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5)
0003          DPMULT=1.134781985
0004          DPI=0.00363138
0005          DO 10 J=1,45
0006          IF(DPI .LT. .0125) GO TO 20
0008          SENS(J)=426.4*(DPI**1.159)
0009          GO TO 10
0010   20     SENS(J)=2.351E12*(DPI**6.262)
0011   10     DPI=DPI*DPMULT
0012          RETURN
0013          END
```

```
                                        THU 16-AUG-79 11:38:54         PAGE 001
FORTRAN IV      V01C-03D                                        ,LP:=EAAMLS.ALL
CORE=08K, UIC=[100,103]
      C      *SENS2.FTN CALCLATES THE POLYDISPERSE SENSITIVITIES FOR
      C      ALL THE THREE WEIGHTINGS FOR THE EAA.
      C
0001         SUBROUTINE SENS2
0002         COMMON SCHISQ,FCHISQ,NCOMP,FNUM,FSUR,FVOL,
            1ANMD1,ANMD2,ASMD1,ASMD2,AVMD1,AVMD2,SIG1,SIG2,
            2DNUMP(10),DSURP(10),DVOLP(10),DNUMM(10),
            3TNUM,TSUR,TVOL,CALIB(11,45),SENS(45),SENSS(10),
            4DPIS(10),SDS(11),DAT(11),TDAT,
            5MASK(5),X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5)
      C
0003         DIMENSION CNUMS1(11),CNUMS2(11),CRNT1(11),CRNT2(11),DUM(11),
            1CNUM1(45),CNUM2(45),PSENS(10)
      C
      C
0004         ALIM=0.0000000001
0005         PI=3.141592653
0006         DO 10 KWEGHT=1,3
0007         GO TO (20,30,40) KWEGHT
0008    20   X1=X(1)
0009         X2=X(3)
0010         GO TO 50
0011    30   X1=EXP(ALOG(X(1))+2.0*(ALOG(X(2)))**2)
0012         X2=EXP(ALOG(X(3))+2.0*(ALOG(X(4)))**2)
0013         GO TO 60
0014    40   X1=EXP(ALOG(X(1))+3.0*(ALOG(X(2)))**2)
0015         X2=EXP(ALOG(X(3))+3.0*(ALOG(X(4)))**2)
0016         GO TO 60
0017    50   CALL CURNT(CNUMS1,CRNT1,X1,X(2),X(5))
0018         FRAC2=1.0-X(5)
0019         CALL CURNT(CNUMS2,CRNT2,X2,X(4),FRAC2)
0020         DO 70 J=1,11
0021         CNUMS1(J)=CNUMS1(J)+CNUMS2(J)
0022    70   CRNT1(J)=CRNT1(J)+CRNT2(J)
0023         DO 80 J=1,10
0024         IF(CRNT1(J) .LT. ALIM .OR. CNUMS1(J) .LT. ALIM) GO TO 90
0026         PSENS(J)=(CNUMS1(J)/CRNT1(J))*1.0E6
0027         GO TO 80
0028    90   PSENS(J)=0.0
0029    80   CONTINUE
0030         GO TO 100
0031    60   CRC1=EXP((-(ALOG(X(1))**2)+(ALOG(X1))**2)/(2.0*(ALOG(X(2))**2)))
0032         CRC2=EXP((-(ALOG(X(3))**2)+(ALOG(X2))**2)/(2.0*(ALOG(X(4))**2)))
0033         WFRAC1=(X(5)*CRC1)/(X(5)*CRC1+(1.0-X(5))*CRC2)
0034         WFRAC2=1.0-WFRAC1
0035         CALL CNUMS(CNUM1,X1,X(2),X(5))
0036         CALL CNUMS(CNUM2,X2,X(4),FRAC2)
0037         CALL DCNVT(CNUM1,CNUMS1)
0038         CALL DCNVT(CNUM2,CNUMS2)
0039         CALL CURNT(DUM,CRNT1,X(1),X(2),X(5))
0040         CALL CURNT(DUM,CRNT2,X(3),X(4),FRAC2)
0041         DO 110 J=1,10
0042         CNUMS1(J)=CRC1*CNUMS1(J)+CRC2*CNUMS2(J)
0043         CRNT1(J)=CRNT1(J)+CRNT2(J)
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:38:54         PAGE 002
CORE=08K, UIC=[100,103]                                   ,LP:=EAAWLS.ALL

0044            IF(CNUMB1(J) .LT. ALIM .OR. CRNT1(J) .LT. ALIM) GO TO 120
0046            PSENS(J)=((PI*CNUMB1(J))/(CRNT1(J)))*1.0E6
0047            IF(KWEGHT .EQ. 3) PSENS(J)=PSENS(J)/6.0
0049            GO TO 110
0050      120   PSENS(J)=0.0
0051      110   CONTINUE
0052      100   GO TO (130,140,150) KWEGHT
0053      130   DO 160 J=1,10
0054      160   DNUMP(J)=DAT(J)*PSENS(J)*4.0
0055            FNUM=X(5)
0056            ANMD1=X1
0057            ANMD2=X2
0058            SIG1=X(2)
0059            SIG2=X(4)
0060            TNUM=0.0
0061            DO 170 J=1,10
0062      170   TNUM=TNUM+DNUMP(J)
0063            TNUM=TNUM/4.0
0064            GO TO 10
0065      140   DO 180 J=1,10
0066      180   DSURF(J)=DAT(J)*PSENS(J)*4.0
0067            ASMD1=X1
0068            ASMD2=X2
0069            FSUR=WFRAC1
0070            TSUR=0.0
0071            DO 190 J=1,10
0072      190   TSUR=TSUR+DSURF(J)
0073            TSUR=TSUR/4.0
0074            GO TO 10
0075      150   DO 200 J=1,10
0076      200   DVOLP(J)=DAT(J)*PSENS(J)*4.0
0077            AVMD1=X1
0078            AVMD2=X2
0079            FVOL=WFRAC1
0080            TVOL=0.0
0081            DO 210 J=1,10
0082      210   TVOL=TVOL+DVOLP(J)
0083            TVOL=TVOL/4.0
0084      10    CONTINUE
0085            RETURN
0086            END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:39:35          PAGE 001
CORE=08K, UIC=[100,103]                                   ,LP:=EAAULS.ALL

0001          SUBROUTINE STEPIT
        C
        C  MINIMIZES A PIECEWISE CONTINUOUS FUNCTION SUBJECT TO ARBITRARY
        C  CONSTRAINTS OF INEQUALITY.
        C  J. A. NELDER AND R. MEAD, THE COMPUTER JOURNAL 7 (1965) 308
        C
        C
        C
0002          COMMON SCHISQ,FCHISQ,NCOMP,FNUM,FSUR,FVOL,
             1ANMD1,ANMD2,ASMD1,ASMD2,AVMD1,AVMD2,SIG1,SIG2,
             2DNUMP(10),DSURP(10),DVOLP(10),DNUMM(10),
             3TNUM,TSUR,TVOL,CALIB(11,45),SENS(45),SENSS(10),
             4DPIS(10),SDS(11),DAT(11),TDAT,
             5MASK(5),X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5)
0003          DIMENSION CHI(6),Z(6,5),ZBAR(5),ZSTAR(5)
        C
0004          NFLAT=1
0005          CHISQ=0.0
0006          NV=5
0007          NFMAX=500
0008       40 HUGE=0.5E 38
0009          ALPHA=1.0
0010          BETA=0.5
0011          GAMMA=2.0
        C
0012       30 DO 130 J=1,NV
0013          IF(MASK(J))130,60,130
0014       60 IF((X(J)+DELTAX(J))-X(J))100,70,100
0015       70 IF(X(J))90,80,90
0016       80 DELTAX(J)=0.01
0017          GO TO 100
0018       90 DELTAX(J)=0.01*X(J)
0019      100 IF(XMAX(J)-XMIN(J))110,110,120
0020      110 XMAX(J)=HUGE
0021          XMIN(J)=-HUGE
0022      120 X(J)=AMAX1(XMIN(J),AMIN1(XMAX(J),X(J)))
0023      130 CONTINUE
        C
        C
        C  CALCULATE INITIAL P(I) AND Y(I).
        C
0024      220 NVA=0
0025          NCOMP=0
0026          J=1
0027      230 IF(MASK(J))270,240,270
0028      240 NVA=NVA+1
0029          DO 250 K=1,NV
0030      250 Z(NVA,K)=X(K)
0031          Z(NVA,J)=Z(NVA,J)+DELTAX(J)
0032          XS=X(J)
0033          X(J)=Z(NVA,J)
0034          CALL OBJCT(NV,CHISQ)
0035      260 X(J)=XS
0036          CHI(NVA)=CHISQ
```

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FORTRAN IV      V01C-03D        THU 14-AUG-79 11:39:35         PAGE`002
CORE=08K, UIC=[100,103]                                     ,LP:=EAANLS.ALL
.
0037      270 J=J+1
0038          IF(J-NV)230,230,280
0039      280 IF(NVA)290,290,310
0040      290 DO 300 J=1,NV
0041      300 MASK(J)=0
0042          GO TO 30
0043      310 NVP=NVA+1
0044          DO 320 J=1,NV
0045      320 Z(NVP,J)=X(J)
        C
        C
0046          CALL OBJCT(NV,CHISQ)
0047      390 CHI(NVP)=CHISQ
0048          BCHISQ=CHISQ
0049      440 JL=NVP
        C
        C  DETERMINE H AND L.
        C
0050      450 JH=JL
0051          DO 490 J=1,NVP
0052          IF(CHI(J)-CHI(JH))470,470,460
0053      460 JH=J
0054      470 IF(CHI(J)-CHI(JL))480,490,490
0055      480 JL=J
0056      490 CONTINUE
0057          IF(JH-JL)560,500,560
0058      500 IF(NFLAT)540,540,1200
0059      540 JH=1
0060          IF(JL-1)560,550,560
0061      550 JH=2
        C
        C  CALCULATE PBAR.
        C
0062      560 DO 600 J=1,NV
0063          IF(MASK(J))600,570,600
0064      570 ZBAR(J)=0.0
0063          DO 590 K=1,NVP
0066          IF(K-JH)580,590,580
0067      580 ZBAR(J)=ZBAR(J)+Z(K,J)
0068      590 CONTINUE
0069          ZBAR(J)=ZBAR(J)/FLOAT(NVA)
0070      600 CONTINUE
        C
        C  ATTEMPT A REFLECTION.
        C  FORM P* .
        C
0071          DO 620 J=1,NV
0072          IF(MASK(J))620,610,620
0073      610 X(J)=(1.0+ALPHA)*ZBAR(J)-ALPHA*Z(JH,J)
0074      620 ZSTAR(J)=X(J)
0075          CALL OBJCT(NV,CHISQ)
0076      630 CHISTR=CHISQ
0077          IF(CHISQ-CHI(JL))690,730,730
        C
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:39:33          PAGE 003
CORE=08K, UIC=[100,103]                                     ,LP:=EAAMLS.ALL

      C  THE REFLECTION SUCCEEDED.  ATTEMPT AN EXPANSION.
      C  FORM P** .
      C
        690 DO 710 J=1,NV
            IF(MASK(J))710,700,710
        700 X(J)=GAMMA*X(J)+(1.0-GAMMA)*ZBAR(J)
        710 CONTINUE
            CALL OBJCT(NV,CHISQ)
        720 IF(CHISQ-CHISTR)1100,790,790
      C
      C  THE REFLECTION FAILED.
      C
        730 DO 750 J=1,NVP
            IF(J-JH)740,750,740
        740 IF(CHISQ-CHI(J))790,750,750
        750 CONTINUE
            IF(CHISQ-CHI(JH))810,830,830
      C
      C
      C  REPLACE P(JH) BY P* .
      C
        790 DO 800 J=1,NV
        800 Z(JH,J)=ZSTAR(J)
            CHI(JH)=CHISTR
            GO TO 1120
      C
        810 DO 820 J=1,NV
        820 Z(JH,J)=X(J)
            CHI(JH)=CHISQ
      C
      C  ATTEMPT A CONTRACTION.
      C  FORM P** .
      C
        830 DO 850 J=1,NV
            IF(MASK(J))850,840,850
        840 X(J)=BETA*Z(JH,J)+(1.0-BETA)*ZBAR(J)
        850 CONTINUE
            CALL OBJCT(NV,CHISQ)
        860 IF(CHISQ-CHI(JH))1100,960,960
      C
      C  THE CONTRACTION FAILED.
      C  REPLACE ALL P(I) BY (P(I)+P(JL))/2.0 .  GUARDING THE ROUNDING.
      C
        960 JS=JL
            DO 1060 J=1,NVP
            IF(J-JL)970,1060,970
        970 DO 1020 K=1,NV
            IF(MASK(K))1020,980,1020
        980 IF(Z(J,K)-Z(JL,K))990,1000,990
        990 X(K)=(Z(J,K)+Z(JL,K))/2.0
            IF(X(K)-Z(J,K))1010,1000,1010
       1000 X(K)=Z(JL,K)
       1010 Z(J,K)=X(K)
       1020 CONTINUE
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:39:35        PAGE 004
CORE=08K, UIC=[100,102]                                  ,LP:=EAAMLB.ALL

0113          CALL OBJCT(NV,CHISQ)
0114    1030 IF(CHISQ-CHI(JS))1040,1050,1050
0115    1040 JS=J
0116    1050 CHI(J)=CHISQ
0117    1060 CONTINUE
0118          JL=JS
0119          GO TO 1120
       C
       C
       C  REPLACE P(JH) BY P* OR P** .
       C
0120    1100 DO 1110 J=1,NV
0121    1110 Z(JH,J)=X(J)
0122          CHI(JH)=CHISQ
       C
       C  TEST FOR CONVERGENCE.
       C
0123    1120 DO 1150 J=1,NV
0124          IF(MASK(J))1150,1130,1150
0125    1130 ZMAX=Z(1,J)
0126          ZMIN=Z(1,J)
0127          DO 1140 K=2,NVP
0128          ZMAX=AMAX1(ZMAX,Z(K,J))
0129    1140 ZMIN=AMIN1(ZMIN,Z(K,J))
0130          IF((ZMAX-ZMIN)-ABS(DELMIN(J)))1150,1150,1260
0131    1150 CONTINUE
       C
0132   1180  IF(CHI(JH)-CHI(JL))1190,1200,1200
0133   1190  JL=JH
0134    1200 DO 1210 J=1,NV
0135    1210 X(J)=Z(JL,J)
0136          CALL OBJCT(NV,CHISQ)
0137          FCHISQ=CHISQ
0138    1250 RETURN
       C
0139   1260  CONTINUE
0140    1270 IF(NCOMP-NFMAX)450,450,1180
0141          END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:40:42          PAGE 001
CORE=08K, UIC=[100,103]                                  ,LP:=EAAWLS.ALL

      C      *WLS.FTN IS THE  WEIGHTED LS FUNCTION TO BE USED
      C      AS THE OBJECTIVE FUNCTION.
      C
0001         SUBROUTINE OBJCT(NV,CHISQ)
0002         COMMON BCHISQ,FCHISQ,NCOMP,FNUM,FSUR,FVOL,
            1AMMO1,AMMO2,ASMO1,ASMO2,AVMO1,AVMO2,SIG1,SIG2,
            2DNUMP(10),DSURP(10),DVOLP(10),DNUMM(10),
            3TNUM,TSUR,TVOL,CALIB(11,43),SENS(43),SENSS(10),
            4DPIS(10),SDS(11),DAT(11),TDAT,
            5MASK(5),X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5)
      C
0003         DIMENSION CRNT1(11),CRNT2(11),DUM(11)
      C
0004         DO 10 J=1,NV
0005         IF(MASK(J)) 10,20,10
0006  20     IF(X(J)-XMAX(J))30,30,40
0007  30     IF(X(J)-XMIN(J))40,10,10
0008  10     CONTINUE
0009         CALL CURNT(DUM,CRNT1,X(1),X(2),X(5))
0010         FRAC2=1.0-X(5)
0011         CALL CURNT(DUM,CRNT2,X(3),X(4),FRAC2)
0012         TCRNT=0.0
0013         DO 60 J=1,11
0014         CRNT1(J)=CRNT1(J)+CRNT2(J)
0015  60     TCRNT=TCRNT+CRNT1(J)
0016         DO 70 J=1,11
0017  70     CRNT1(J)=CRNT1(J)/TCRNT
0018         CHISQ=0.0
0019         DO 80 J=1,11
0020         AF=DAT(J)-CRNT1(J)
0021         IF( ABS(AF) .LT. .001) GO TO 80
0023         IF( DAT(J) .LT. .0001) GO TO 90
0025         CHISQ=CHISQ+(AF**2)/DAT(J)
0026         GO TO 80
0027  90     CHISQ=CHISQ+10
0028  80     CONTINUE
0029         NCOMP=NCOMP+1
0030         RETURN
0031  40     CHISQ=1.0E20
0032         RETURN
0033         END
```

```
FORTRAN IV      V01C-03D         THU 16-AUG-79 11:41:06            PAGE 001
CORE=08K, UIC=[100,103]                                    ,LP:=EAAALS.ALL

      C     *CURNT.FTN CALCULATES THE SIMULATED CURRENT DUE TO
      C     A UNIMODAL AEROSOL SIZE DISTRIBUTION FOR THE EAA.
      C
      C
0001        SUBROUTINE CURNT(CNUMS1,CRNT,ANMD,SIGMA,FRAC1)
      C
0002        COMMON SCHISQ,FCHISQ,NCOMP,FNUM,FSUR,FVOL,
           1ANMD1,ANMD2,ASMD1,ASMD2,AVMD1,AVMD2,SIG1,SIG2,
           2DNUMP(10),DSURP(10),DVOLP(10),DNUMM(10),
           3TNUM,TSUR,TVOL,CALIB(11,45),SENS(45),SENSS(10),
           4DPIS(10),SDS(11),DAT(11),TDAT,
           5MASK(5),X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5)
      C
0003        DIMENSION CNUM(45),CRNT(11),CNUMS1(11)
0004        CALL CNUMS(CNUM,ANMD,SIGMA,FRAC1)
0005        DO 10 J=1,11
0006        CRNT(J)=0.0
0007        DO 10 I=1,45
0008  10    CRNT(J)=CRNT(J)+CALIB(J,I)*SENS(I)*CNUM(I)
0009        CALL DCNVT(CNUM,CNUMS1)
0010        RETURN
0011        END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:41:25        PAGE 001
CORE=08K, UIC=[100,103]                                    ,LP:=ERRMAT.ALL

        C       ERRMAT.FTN CALCULATES THE CURVATURE MATRIX FOR
        C       FOR THE OBJECTIVE FUNCTION
        C
0001            SUBROUTINE ERRMAT(ALPHA,MODE)
0002            COMMON SCHISQ,FCHISQ,NCOMP,FNUM,FSUR,FVOL,
               1AMMO1,AMMO2,ASMO1,ASMO2,AVMO1,AVMO2,SIG1,SIG2,
               2DNUMP(10),DSURP(10),DVOLP(10),DNUMM(10),
               3TNUM,TSUR,TVOL,CALIB(11,45),SENS(45),SENSS(10),
               4DPIS(10),SDS(11),DAT(11),TDAT,
               5MAGN(5),X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5)
        C
0003            DIMENSION ALPHA(5,5),DELTA(5)
0004            DOUBLE PRECISION ALPHA
0005            DELTA(1)=0.7783*X(1)
0006            DELTA(2)=10.**(ALOG10(X(2))*1.25) -X(2)
0007            IF (DELTA(2) .LT. .1) DELTA(2)=.1
0009            DELTA(3)=0.7783*X(3)
0010            DELTA(4)=10.**(ALOG10(X(4))*1.25)-X(4)
0011            IF (DELTA(4) .LT. .1) DELTA(4)=.1
0013            DELTA(5)=X(5)*(1.0-X(5))
0014            IF (DELTA(5) .GT. .1) DELTA(5)=.1
0016            DO 100 J=1,5
0017            XJ=X(J)+DELTA(J)
0018            IF(XMAX(J)-XJ) 120,110,110
0019    120     DELTA(J)=XMAX(J)-X(J)
0020    110     XJ=X(J)-DELTA(J)
0021            IF (XMIN(J)-XJ) 100,100,130
0022    130     DELTA(J)=X(J)-XMIN(J)
0023    100     CONTINUE
0024            IF (MODE .EQ. 1) NV=2
0026            IF(MODE .EQ. 2) NV=5
0028            DO 5 J=1,NV
0029            DO 5 K=1,NV
0030    5       ALPHA(J,K)=0.0
0031            DO 10 J=1,NV
0032            XJ=X(J)
0033            X(J)=X(J)+DELTA(J)
0034            CALL OBJCT (NV,CHISQ2)
0035            ALPHA(J,J)=CHISQ2-2.0*FCHISQ
0036            DO 20 K=1,NV
0037            IF(K-J) 30,20,40
0038    30      ALPHA(K,J)=(ALPHA(K,J)-CHISQ2)/2.0
0039            ALPHA(J,K)=ALPHA(K,J)
0040            GOTO 20
0041    40      ALPHA(J,K)=FCHISQ-CHISQ2
0042            XK=X(K)
0043            X(K)=X(K)+DELTA(K)
0044            CALL OBJCT(NV,CHISQ3)
0045            ALPHA(J,K)=ALPHA(J,K)+CHISQ3
0046            X(K)=XK
0047    20      CONTINUE
0048            X(J)=XJ-DELTA(J)
0049            CALL OBJCT (NV,CHISQ3)
0050            X(J)=XJ
```

```
FORTRAN IV      V01C-03D       THU 16-AUG-79 11:41:25          PAGE 002
CORE=08K, UIC=[100,103]                                   ,LP:=EAAHLS.ALL

            ALPHA(J,J)=(ALPHA(J,J)+CHISQ2)/2.0
      10    CONTINUE
            DO 30 J=1,NV
            DO 30 K=1,NV
            ALPHA(J,K)=ALPHA(J,K)/(DELTA(J)*DELTA(K))
      30    CONTINUE
            CALL MATINV (ALPHA, NV,DET)
      80    RETURN
            END
```

```
FORTRAN IV      V01C-03D         THU 16-AUG-79 11:41:56          PAGE 001
CORE=08K, UIC=[100,103]                                   .LP:=EAAMLB.ALL

      C
      C     SUBROUTINE MATINV INVERTIS A SYMMETRIC MATRIX AND CALCULATES
      C     ITS DETERMINANT
      C
      C     FROM BEVINGTON BASED ON IBM SCIENTIFIC SUBROUTINE PACKAGE
      C
      C
      C
0001        SUBROUTINE MATINV(ARRAY,NORDER,DET)
0002        DOUBLE PRECISION ARRAY, AMAX, SAVE
0003        DIMENSION ARRAY(5,5),IK(5),JK(5)
      C     WRITE(6,250) ARRAY
      C250  FORMAT (5(3X,1PE10.3))
0004  10    DET=1.0
0005  11    DO 100 K=1,NORDER
      C
      C     FIND THE LARGEST ELEMENT ARRAY(I,J) IN REST OF MATRIX
      C
0006        AMAX=0.0
0007  21    DO 30 I=K,NORDER
0008        DO 30 J=K,NORDER
0009  23    IF (DABS(AMAX) -DABS( ARRAY (I,J))) 24,24,30
0010  24    AMAX=ARRAY(I,J)
0011        IK(K)=I
0012        JK(K)=J
      C     WRITE(6,150) ARRAY(I,J),AMAX,K,I,J
      C150  FORMAT(5X,'30',2(3X,1PE10.3),3(5X,0PI3))
0013  30    CONTINUE
      C
      C     INTERCHANGE ROWS AND COLUMNS TO PUT AMAX IN ARRRAY(K,K)
      C
0014  31    IF (AMAX) 41,32,41
0015  32    DET=0.0
0016        GO TO 140
0017  41    I=IK(K)
0018        IF (I-K) 21,51,43
0019  43    DO 50 J=1,NORDER
0020        SAVE= ARRAY(K,J)
0021        ARRAY(K,J)= ARRAY(I,J)
0022  50    ARRAY(I,J)=-SAVE
      C     WRITE (6,220) ARRAY(K,J),SAVE,K,I,J
      C220  FORMAT(5X,'31',2(3X,1PE10.3),3(5X,0PI3))
0023  51    J=JK(K)
0024        IF (J-K) 21,61,53
0025  53    DO 60 I=1,NORDER
0026        SAVE=ARRAY(I,K)
0027        ARRAY(I,K)=ARRAY(I,J)
0028  60    ARRAY(I,J)=-SAVE
      C     WRITE(6,230) ARRAY(I,K),SAVE,K,I,J
      C230  FORMAT(5X,'61',2(3X,1PE10.3),3(5X,0PI3))
      C
      C     ACCUMULATE ELEMENTS OF INVERSE MATRIX
      C
0029  61    DO 70 I=1,NORDER
0030        IF (I-K) 63,70,63
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:41:56          PAGE 002
CORE=08K, UIC=[100,103]                                    ,LP:=EAAMLS.ALL

0031  63     ARRAY(I,K)=-ARRAY(I,K)/AMAX
      C      WRITE(6,160) ARRAY(I,K),AMAX,K,I
      C160   FORMAT(3X,'70',2(3X,1PE10.3),2(5X,0PI3))
0032  70     CONTINUE
0033  71     DO 80 I=1,NORDER
0034         DO 80 J=1,NORDER
0035         IF (I-K) 74,80,74
0036  74     IF (J-K) 75,80,75
0037  75     ARRAY(I,J)=ARRAY(I,J)+ARRAY(I,K)*ARRAY(K,J)
      C      WRITE(6,170) ARRAY(I,J),ARRAY(I,K),ARRAY(K,J),K,I,J
      C170   FORMAT(3X,'80',3(3X,1PE10.3),3(5X,0PI3))
0038  80     CONTINUE
0039  81     DO 90 J=1,NORDER
0040         IF (J-K) 83,90,83
0041  83     ARRAY(K,J)=ARRAY(K,J)/AMAX
      C      WRITE(6,180) ARRAY(K,J),AMAX,K,J
      C180   FORMAT(3X,'90',2(3X,1PE10.3),2(5X,0PI3))
0042  90     CONTINUE
0043         ARRAY(K,K)=1.0/AMAX
      C      WRITE(6,190) ARRAY(K,K),AMAX,K
      C190   FORMAT(4X,'100',2(3X,1PE10.3),5X,0PI3)
0044  100    DET=DET*AMAX
      C
      C      RESTORE ORDERING OF MATRIX
      C
0045  101    DO 130 L=1,NORDER
0046         K=NORDER-L+1
0047         J=IK(K)
0048         IF (J-K) 111,111,105
0049  105    DO 110 I=1,NORDER
0050         SAVE=ARRAY(I,K)
0051         ARRAY(I,K)=-ARRAY(I,J)
      C      WRITE(6,200)ARRAY(I,J),SAVE,K,I,J
      C200   FORMAT(3X,'110',2(3X,1PE10.3),3(5X,0PI3))
0052  110    ARRAY(I,J)=SAVE
0053  111    I=JK(K)
0054         IF(I-K) 130,130,113
0055  113    DO 120 J=1,NORDER
0056         SAVE=ARRAY(K,J)
0057         ARRAY(K,J)=-ARRAY(I,J)
      C      WRITE(6,210) ARRAY(I,J),SAVE,K,I,J
      C210   FORMAT(4X,'120',2(3X,1PE10.3),3(5X,0PI3))
0058  120    ARRAY(I,J)=SAVE
0059  130    CONTINUE
0060  140    RETURN
0061         END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:42:25          PAGE 001
CORE=08K, UIC=[100,103]                                     ,LP:=EAAHLS.ALL
      C       THIS SUBROUTINE CALCULATES THE NUMBER IN THE 45 DIVISIONS
      C
              SUBROUTINE CNUMS(CNUM,ANMD,SIGMA,FRAC)
              DIMENSION CNUM(45)
             BD1=0.003398212
              DPMULT=1.154781985
              XANMD=ALOG(ANMD)
              XSIGMA=ALOG(SIGMA)
              DO 10 J=1,45
              A=(ALOG(BD1)-XANMD)/XSIGMA
              B=(ALOG(BD1*DPMULT)-XANMD)/XSIGMA
              BD1=BD1*DPMULT
      10      CNUM(J)=APROX(A,B)*FRAC
              RETURN
              END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:42:36         PAGE 001
CORE=08K, UIC=[100,103]                                       ,LP:=EAAMLS.ALL

      C       THIS IS AN APPROXIMATION FOR THE CUMULATIVE
      C       NORMAL DISTRIBUTION FUNCTION. REF. ABRAMOWITZ
      C       AND STEGUN: MATHEMATICAL HANDBOOK.
0001          FUNCTION APROX(A,B)
0002          X1=ABS(A)
0003          X2=ABS(B)
0004          IF(X1) 10,20,10
0005    10     PX1=P(X1)
0006          GO TO 30
0007    20     PX1=0.5
0008    30     IF(X2) 35,40,35
0009    35     PX2=P(X2)
0010          GO TO 45
0011    40     PX2=0.5
0012    45     SIN=A*B
0013          IF(SIN)60,50,50
0014    50     APROX=ABS(PX2-PX1)
0015          GO TO 70
0016    60     APROX=1.0-(PX2+PX1)
0017    70     RETURN
0018          END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:42:48          PAGE 001
CORE=08K, UIC=[100,103]                                    ,LP:=EAAMLS.ALL

        C       THIS FUNCTION IS USED WITH APROX
0001            FUNCTION P(X)
0002            DIMENSION B(6)
0003            DATA B/0.319381530,-0.356563782,1.781477937,-1.821255978,
              11.330274429,0.2316419/
0004            T=1.0/(1.0+B(6)*X)
0005            P=Z(X)*(B(1)*T+B(2)*T**2+B(3)*T**3+B(4)*T**4+B(5)*T**5)
0006            RETURN
0007            END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:42:56          PAGE 001
CORE=08K, UIC=[100,103]                                   .LP:=EAAMLS.ALL

      C        THIS FUNCTION IS USED WITH APPROX
               FUNCTION Z(X)
               PI=3.1415927
               S2PI=SQRT(2.0*PI)
               ARG=X**2/2.0
               IF(ARG-87.0) 10,20,20
      10        A=EXP(ARG)
               Z=1.0/(S2PI*A)
               RETURN
      20        Z=0.0
               RETURN
               END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:43:05          PAGE 001
CORE=08K, UIC=[100,103]                                    ,LP:=EAAMLS.ALL

      C       *BLOCK.DAT CONTAINS THE EAA RESPONSE MATRIX,SENSITIVITIES
      C       BOUNDS,AND MID. POINT DIAMETERS OF THE EAA CHANNELS.
      C       IT ALSO CONTAINS THE VALUES OF THE CONSTANTS FOR
      C       USING THE SIMPLEX METHOD.
      C
0001          BLOCK DATA
0002          COMMON BCHISQ,FCHISQ,NCOMP,FNUM,FSUR,FVOL,
             1ANMD1,ANMD2,ASMD1,ASMD2,AVMD1,AVMD2,SIG1,SIG2,
             2DNUMP(10),DSURP(10),DVOLP(10),DNUMM(10),
             3TNUM,TSUR,TVOL,CALIB(11,43),SENS(43),SENS3(10),
             4DPIS(10),BDS(11),DAT(11),TDAT,
             5MASK(5),X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5)
0003          DATA CALIB/
             1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
             1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
             1 1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .500,.500,.000,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.500,.500,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,.480,.320,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,.050,.930,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,.080,.920,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,.000,1.00,.000,.000,.000,.000,.000,.000,.000,
             1 .000,.000,.000,.310,.480,.010,.000,.000,.000,.000,.000,
             1 .000,.000,.000,.440,.520,.040,.000,.000,.000,.000,.000,
             1 .000,.000,.000,.490,.070,.440,.000,.000,.000,.000,.000,
             1 .000,.000,.000,.150,.480,.195,.175,.000,.000,.000,.000,
             1 .000,.000,.000,.145,.355,.325,.175,.000,.000,.000,.000,
             1 .000,.000,.000,.035,.345,.315,.085,.020,.000,.000,.000,
             1 .000,.000,.000,.000,.140,.650,.170,.020,.020,.000,.000,
             1 .000,.000,.000,.000,.050,.610,.310,.030,.000,.000,.000,
             1 .000,.000,.000,.000,.000,.510,.405,.085,.000,.000,.000,
             1 .000,.000,.000,.000,.000,.450,.480,.070,.000,.000,.000,
             1 .000,.000,.000,.000,.000,.400,.510,.090,.000,.000,.000,
             1 .000,.000,.000,.000,.000,.130,.670,.170,.030,.000,.000,
             1 .000,.000,.000,.000,.000,.030,.470,.380,.080,.040,.000,
             1 .000,.000,.000,.000,.000,.000,.350,.460,.140,.040,.010,
             1 .000,.000,.000,.000,.000,.000,.220,.480,.200,.060,.040,
             1 .000,.000,.000,.000,.000,.000,.150,.450,.250,.090,.060,
             1 .000,.000,.000,.000,.000,.000,.080,.420,.320,.110,.070,
             1 .000,.000,.000,.000,.000,.000,.020,.320,.380,.180,.100,
             1 .000,.000,.000,.000,.000,.000,.000,.310,.360,.180,.150,
             1 .000,.000,.000,.000,.000,.000,.000,.200,.390,.210,.200,
             1 .000,.000,.000,.000,.000,.000,.000,.195,.305,.260,.240,
             1 .000,.000,.000,.000,.000,.000,.000,.130,.270,.250,.350,
             1 .000,.000,.000,.000,.000,.000,.000,.110,.280,.250,.360,
             1 .000,.000,.000,.000,.000,.000,.000,.070,.230,.250,.450,
             1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
             1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.500,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.300,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.300,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.300,
           1 .000,.000,.000,.000,.000,.000,.000,.040,.200,.260,.300/
0004         DATA SENSS/0.0,0.105,2.4,6.0,11.3,22.5,41.9,81.0,150.,283./
0005         DATA DELTAX/.005,.13,.1,.13,.1/
0006         DATA DELMIN/.001,.07,.05,.07,.05/
0007         DATA DPIS/.0042,.0075,.0133,.0237,.0422,
           1.075,.133,.237,.422,.75/
0008         DATA SDS/0.0032,.0056,.01,.0178,.0316,.0562,
           1.1,.178,.316,.562,1.0/
0009         DATA XMIN/0.001,1.05,.001,1.05,0.0/
0010         DATA XMAX/1.0,3.0,1.0,3.0,1.0/
0011         END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:43:36            PAGE 001
CORE=08K, UIC=[100,103]                                      ,LP:=EAAHLS.ALL

      C      *DCNVT.FTN CONVERTS THE DETAIL 45 DIVISION NUMBER
      C      DISTRIBUTION TO THE 11 CHANNELS CORRESPONDING TO
      C      THE EAA.
      C
      C
0001         SUBROUTINE DCNVT(CNUM,CNUMS)
0002         DIMENSION CNUM(45),CNUMS(11)
0003         CNUMS(1)=CNUM(1)+CNUM(2)+CNUM(3)+0. 5*CNUM(4)
0004         I=1
0005         DO 10 J=6,42,4
0006         I=I+1
0007  10     CNUMS(I)=0. 5*CNUM(J-2)+CNUM(J-1)+CNUM(J)
            1+CNUM(J+1)+0. 5*CNUM(J+2)
0008         CNUMS(11)=0. 5*CNUM(40)+CNUM(41)+CNUM(42)+CNUM(43)+CNUM(44)
            1+CNUM(45)
0009         RETURN
0010         END
```

APPENDIX E

COMPUTER PROGRAM FOR DB-CNC DATA REDUCTION
USING TWOMEY'S NON-LINEAR ITERATIVE ALGORITHM
(DIBTIA)

```
FORTRAN IV      V01C-03D       THU 16-AUG-79 11:53:03          PAGE 001
CORE=08K, UIC=[100,100]                                    ,LP:=DISTIA.ALL

        C       *DISTIA.FTN
        C       DISTIA.FTN USES TWOMEY'S NON-LINEAR
        C       ALGORITHM FOR REDUCING DIFFUSION BATTERY
        C       DATA. THIS PROGRAM CAN BE USED FOR THE TSI
        C       SCREEN BATTERY FOR 4.0 OR 6.0 LPM FLOW RATE OR FOR THE
        C       COLLIMATED HOLE STRUCTURE(CHS) BATTERY FOR ANY FLOW.
        C       THE ITERATIVE PROCEDURE IS STOPPED EITHER
        C       WHEN WLS IS LESS THEN 0.001 OR THE NUMBER
        C       OF ITRATIONS IS GREATER THAN 100. WHICHEVER OCCURS FIRST
        C       AFTER THE FIRST 20 ITERATIONS.
        C
        C
        C
        C
0001            COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
               1CLOSS(45),ITER,FLOW,DIA(45),X
0002            DIMENSION TITTLE(10)
0003            CALL ASSIGN(6,'LP:')
0004            DIA(1)=0.00316228
0005            DPMULT=1.154781985
0006            DO 5 J=2,45
0007    5       DIA(J)=DIA(J-1)*DPMULT
0008    230     WRITE (5,10)
0009    10      FORMAT(3X,'ENTER DATA DESCRIPTION (40 CHARACTERS MAXM.)')
0010            READ(5,20) TITTLE
0011    20      FORMAT(10A4)
        C
        C       SET UP THE LOSS MATRIX
        C
0012    50      WRITE(5,30)
0013    30      FORMAT(3X,'ENTER',/5X,'1 IF TSI SCREEN BATTERY WAS USED',/
               13X,'OR 2 IF CHS BATTERY WAS USED',/)
0014            READ(5,40) IBAT
0015    40      FORMAT(I1)
0016            IF(IBAT .EQ. 1 .OR. IBAT .EQ. 2) GO TO 45
0018            GO TO 50
0019    45      IF(IBAT .EQ. 1) CALL TSI
0021            IF(IBAT .EQ. 2) CALL CHS
        C
        C       ENTER DATA
        C
0023            WRITE(5,60)
0024            DO 65 J=1,12
0025    65      DAT(J)=0.0
0026    60      FORMAT(3X,'ENTER THE DATA FROM CNC',/)
0027            IF (IBAT .EQ. 1) KDAT=10
0029            IF (IBAT .EQ. 2) KDAT=11
0031            READ(5,70) TDAT,(DAT(I),I=1,KDAT)
0032    70      FORMAT(12F10.0)
0033            KDAT=KDAT+1
0034            KDAT1=KDAT-1
0035            DO 80 J=1,KDAT1
0036            NLAST=KDAT-J+1
0037            DAT(NLAST)=DAT(NLAST-1)-DAT(NLAST)
0038    80      DAT(NLAST)=AMAX1(DAT(NLAST),0.0)
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:53:03          PAGE 002
CORE=08K, UIC=[100,103]                                    ,LP:=DISTIA.ALL
0039          DAT(I)=AMAX1((TDAT-DAT(I)),0.0)
        D     WRITE(6,101) TDAT,DAT,KDAT
        D101  FORMAT(3X,'INPUT DATA',/13(X,1PE9.3),/3X,I4)
0040          CALL DIFFTWO
        C
        C     CALCULATE VALUES FOR OUTPUT
        C
0041          WRITE(6,90) TITTLE,ITER,X
0042    90    FORMAT(///20X,'2/21/79 VERSION'///9X,10A4//10X,
             1'NO. OF ITERATIONS= ',I3,
             15X,'CHISQ=',1PE10.3)
0043          DO 100 J=3,39,4
0044    100   CLOSS(J)=(0.5*CLOSS(J-2)+CLOSS(J-1)+CLOSS(J)+
             1CLOSS(J+1)+0.5*CLOSS(J+2))/4.0
0045          DO 110 J=3,43,4
0046          CLOSS(J+1)=CLOSS(J)*3.14*(DIA(J)**2)
0047    110   CLOSS(J+2)=CLOSS(J)*0.523*(DIA(J)**3)
0048          THUM=0.0
0049          TSURF=0.0
0050          TVOL=0.0
0051          DO 120 J=3,43,4
0052          THUM=THUM+CLOSS(J)
0053          TSURF=TSURF+CLOSS(J+1)
0054    120   TVOL=TVOL+CLOSS(J+2)
0055          WRITE(6,130)
0056    130   FORMAT(3X,'PORT NO.',3X,'OBS. DATA',3X,'CALC. DATA',
             13X,'MID PT. DIA',4X,'DNDLGD',5X,'DSDLGD',3X,'DVDLGD',/)
0057          I=0
0058          DO 150 J=3,39,4
0059          I=I+1
0060    150   WRITE(6,160) I,DAT(I),CDAT(I),DIA(J),CLOSS(J),
             1CLOSS(J+1),CLOSS(J+2)
0061    160   FORMAT(3X,I2,10X,F7.4,2X,F7.4,7X,F6.4,3(4X,1PE9.3))
0062          I=I+1
0063          WRITE(6,160)I,DAT(I),CDAT(I)
0064          GO TO (170,180) IBAT
0065    180   I=I+1
0066          WRITE(6,160) I,DAT(I),CDAT(I)
0067    170   THUM=THUM/4.0
0068          TSURF=TSURF/4.0
0069          TVOL=TVOL/4.0
0070          WRITE(6,190) THUM,TSURF,TVOL
0071    190   FORMAT(3X,'TOTALS',25X,3(4X,1PE9.3))
0072          DO 195 J=3,39,4
0073    195   CLOSS(J)=CLOSS(J)/(THUM*4.0)
0074          WRITE(6,197)(CLOSS(J),J=3,39,4)
0075    197   FORMAT(//5X,10(4X,F6.4),/)
0076          WRITE(5,200)
0077    200   FORMAT(3X,'ENTER 1 FOR ANOTHER CALC. 2 FOR STOP',/)
0078    205   READ(5,210) IFLAG
0079    210   FORMAT(I1)
0080          IF (IFLAG .LT. 1 .OR. IFLAG .GT. 2) GO TO 205
0082          GO TO (230,220) IFLAG
0083    220   STOP
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:33:03        PAGE 003
CORE=08K, UIC=[100,103]                            ,LP:=DISTIA.ALL

0084        END
```

```
FORTRAN IV      V01C-03B        THU 16-AUG-79 11:22:46        PAGE 001
CORE=08K, UIC=[100,103]                              ,LP:=DIFTIA.ALL

      C      *DIFFTWO.FTN
      C
      C      THIS SUBROUTINE USES TWOMEY'S NONLINEAR ALGORITHM
      C      FOR SOLVING THE DIFFUSION BATTERY EQNS.
      C
      C
0001         SUBROUTINE DIFFTWO
0002         COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
            1CLOSS(45),ITER,FLOW,DIA(45),E
0003         DIMENSION CHCEFF(45)
0004         DATA CHCEFF/45*1.0/
0005         KMAX=20
0006         JMAX=45
0007         DO 10 J=1,JMAX
0008  10     CLOSS(J)=TDAT/JMAX
0009         ITER=0
0010  100    DO 20 K=1,KMAX
0011         DO 30 I=1,KDAT
0012         RATIO=0.0
0013         DO 40 J=1,JMAX
0014  40     RATIO=RATIO+CALIB(I,J)*CLOSS(J)*CHCEFF(J)
0015         IF(RATIO) 30,30,60
0016  60     RATIO=DAT(I)/RATIO-1.0
0017         DO 70 J=1,JMAX
0018  70     CLOSS(J)=CLOSS(J)*(1.0+(RATIO*CALIB(I,J)))
0019  30     CONTINUE
0020  20     CONTINUE
0021         ITER=ITER+KMAX
0022         IF(ITER.GT.100) GO TO 90
0024         DO 80 I=1,KDAT
0025         CDAT(I)=0.0
0026         DO 80 J=1,JMAX
0027  80     CDAT(I)=CDAT(I)+CALIB(I,J)*CLOSS(J)*CHCEFF(J)
0028         CALL OBJECT
0029         IF(E .LT. .005) GO TO 90
0031         KMAX=5
0032         GO TO 100
0033  90     CONTINUE
      D      WRITE(6,630)
      D630   FORMAT(5X,'DETAILED SOLN')
      D      WRITE(6,620) CLOSS
      D620   FORMAT(10(2X,1PE9.2))
0034         RETURN
0035         END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:54:07          PAGE 001
CORE=08K, UIC=[100,103]                                    .LP:=DISTIA.ALL

      C       CHB.FTN
      C
      C       CHB.FTN SETS UP THE LOSS MATRIX
      C       FOR THE COLLIMATED HOLE STRUCTURE BATTERY
      C       FOR ANY SPECIFIED FLOW
      C
      C
0001          SUBROUTINE CHB
0002          COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
             1CLOSS(45),ITER,FLOW,DIA(45),I
0003          DIMENSION ALEN(18),KTAP(11),P(18)
0004          DATA ALEN/2*0.3175,0.635,0.9525,1.27,1.905,
             12.54,1.27,10*2.54/
0005          DATA KTAP/1,2,3,4,5,6,7,9,11,14,18/
0006          DPI=0.00316228*.0001
0007          DPMULT=1.154781985
0008          WRITE(5,10)
0009    10    FORMAT(3X,'ENTER THE FLOW RATE(LPM) (F FORMAT)',/)
0010          READ(5,20) FLOW
0011    20    FORMAT(F13.0)
0012          FLOW=FLOW*16.6667
0013          DO 30 I=1,45
0014          D=(2.37E-11/DPI)*(1.0+(1.306E-5/DPI)*(1.246+
             10.42*EXP(-66616.0*DPI)))
0015          DO 40 J=1,18
0016          AMU=3.1459*ALEN(J)*D*9790./FLOW
0017          P(J)=0.819*EXP(-3.657*AMU)+0.097*EXP(-22.3*AMU)
             1+0.032*EXP(-57.0*AMU)+0.027*EXP(-123.0*AMU)
             2+0.025*EXP(-750.0*AMU)
0018    40    CONTINUE
0019          DO 50 J=2,12
0020          MUL=KTAP(J-1)
0021          CALIB(J-1,I)=1.0
0022          DO 60 KMUL=1,MUL
0023    60    CALIB(J-1,I)=CALIB(J-1,I)*P(KMUL)
0024    50    CONTINUE
0025          DPI=DPI*DPMULT
0026    30    CONTINUE
0027          DO 70 I=1,45
0028          P(1)=1.0-CALIB(1,I)
0029          DO 80 J=2,11
0030          P(J)=CALIB(J-1,I)-CALIB(J,I)
0031    80    CONTINUE
0032          P(12)=CALIB(11,I)
0033          DO 90 J=1,12
0034    90    CALIB(J,I)=P(J)
0035    70    CONTINUE
0036          RETURN
0037          END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:54:30         PAGE 001
CORE=08K, UIC=[100,103]                                     ,LP:=DISTIA.ALL

        C       *TSI.FTN
        C
        C       THIS SUBROUTINE COMPUTES THE LOSS MATRIX FOR
        C       TSI SCREEN BATTERY FOR 4.0 OR 6.0 LPM FLOW RATES.
        C
        C
0001            SUBROUTINE TSI
0002            COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
               1CLOSS(45),ITER,FLOW,DIA(45),X
0003      20    WRITE(5,10)
0004      10    FORMAT(5X,'IS THE FLOW RATE 4.0 OR 6.0 LPM?',2X,
               1'(F FORMAT)',)
0005            READ(5,30) FLOW
0006      30    FORMAT(F13.0)
0007            IF (FLOW .EQ. 4.0 .OR. FLOW .EQ. 6.0 ) GO TO 50
0009            GO TO 20
0010      50    IF(FLOW .EQ. 4.0 ) GO TO 60
0012            AK1=-1.1171981
0013            CK1=-3.067
0014            GO TO 70
0015      60    AK1=-1.14423
0016            CK1=-2.990605
0017      70    DO 90 J=1,45
0018            NSCRN=0
0019            SLOPE=-10.0**(AK1*ALOG10(DIA(J))+CK1)
0020            DO 80 I=2,11
0021            CALIB(I-1,J)=(10.0**(SLOPE*NSCRN))-(10.0**(SLOPE*(NSCRN+I-1)
               1))
0022      80    NSCRN=NSCRN+I-1
0023      90    CALIB(11,J)=10.0**(SLOPE*55.)
0024            RETURN
0025            END
```

```
FORTRAN IV      V01C-03D          THU 16-AUG-79 11:54:47          PAGE 001
CORE=08K, UIC=[100,103]                                       ,LP:=DISTIA.ALL
      C      *MLS.FTN
      C
      C      THIS SUBROUTINE COMPUTES THE CHI-SQUARE FOR
      C      THE OBSERVED AND CALCULATED DATA FOR THE DIFFUSION BATTERY.
      C
      C
0001         SUBROUTINE OBJCT
0002         COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
            1CLOSS(45),ITER,FLOW,DIA(45),X
0003         X=0.0
0004         TCDAT=0.0
0005         DO 5 J=1,KDAT
0006   5     TCDAT=TCDAT+CDAT(J)
0007         DO 10J=1,KDAT
0008         AF=(CDAT(J)/TCDAT)-(DAT(J)/TDAT)
0009         IF(ABS(AF) .LT. .001 .AND. CDAT(J) .LT. .0001) GO TO 10
0011         X=X+(AF**2)/(DAT(J)/TDAT)
0012   10    CONTINUE
0013         RETURN
0014         END
```

APPENDIX F

COMPUTER PROGRAM FOR DB-CNC DATA REDUCTION USING NON-LINEAR ITERATIVE ALGORITHM (II)

(DIBKIA)

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:48:23          PAGE 001
CORE=08K, UIC=[100,103]                                   .LP:=DISKIA.ALL
        C       *DISKIA.FTN
        C       DISKIA.FTN USES A NON-LINEAR [redacted]
        C       ALGORITHM FOR REDUCING DIFFUSION BATTERY
        C       DATA.  THIS PROGRAM CAN BE USED FOR THE TSI
        C       SCREEN BATTERY FOR 4.0 OR 6.0 LPM FLOW RATE OR FOR THE
        C       COLLIMATED HOLE STRUCTURE(CHS) BATTERY FOR ANY FLOW.
        C       THE ITERATIVE PROCEDURE IS STOPPED EITHER
        C       WHEN WLS IS LESS THEN 0.005 OR THE NUMBER
        C       OF ITRATIONS IS GREATER THAN 100, WHICHEVER OCCURS FIRST
        C       AFTER THE FIRST 20 ITERATIONS.
        C
        C
        C
0001            COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
               1CLOSS(45),ITER,FLOW,DIA(45),X
0002            DIMENSION TITTLE(10)
0003            CALL ASSIGN(6,'LP:')
0004            DIA(1)=0.00316228
0005            DPMULT=1.154781985
0006            DO 5 J=2,45
0007    5       DIA(J)=DIA(J-1)*DPMULT
0008    230     WRITE (5,10)
0009    10      FORMAT(5X,'ENTER DATA DESCRIPTION (40 CHARACTERS MAXM.)')
0010            READ(5,20) TITTLE
0011    20      FORMAT(10A4)
        C
        C       SET UP THE LOSS MATRIX
        C
0012    50      WRITE(5,30)
0013    30      FORMAT(5X,'ENTER',/5X,'1 IF TSI SCREEN BATTERY WAS USED',/
               15X,'OR 2 IF CHS BATTERY WAS USED',/)
0014            READ(5,40) IBAT
0015    40      FORMAT(I1)
0016            IF(IBAT .EQ. 1 .OR. IBAT .EQ. 2) GO TO 45
0018            GO TO 50
0019    45      IF(IBAT .EQ. 1) CALL TSI
0021            IF(IBAT .EQ. 2) CALL CHS
        C
        C       ENTER DATA
        C
0023            WRITE(5,60)
0024            DO 65 J=1,12
0025    65      DAT(J)=0.0
0026    60      FORMAT(5X,'ENTER THE DATA FROM CNC',/)
0027            IF (IBAT .EQ. 1) KDAT=10
0029            IF (IBAT .EQ. 2) KDAT=11
0031            READ(5,70) TDAT,(DAT(I),I=1,KDAT)
0032    70      FORMAT(12F10.0)
0033            KDAT=KDAT+1
0034            KDAT1=KDAT-1
0035            DO 80 J=1,KDAT1
0036            KLAST=KDAT-J+1
0037            DAT(KLAST)=DAT(KLAST-1)-DAT(KLAST)
0038    80      DAT(KLAST)=AMAX1(DAT(KLAST),0.0)
```

```
FORTRAN IV      V01C-03D          THU 16-AUG-79 11:48:35            PAGE 002
CORE=08K, UIC=[100,103]
                                                           ,LP:=DISKIA.ALL
0039          DAT(I)=AMAX1((TDAT-DAT(I)),0.0)
        D     WRITE(6,101) TDAT,DAT,KDAT
        D101  FORMAT(3X,'INPUT DATA',/13(X,1PE9.3),/5X,I4)
0040          CALL DIFFTER
        C
        C     CALCULATE VALUES FOR OUTPUT
        C
0041          WRITE(6,90) TITTLE,ITER,X
0042    90    FORMAT(///9X,10A4//10X,
             1'NO. OF ITERATIONS= ',I3,
             15X,'CHISQ=',1PE10.3)
0043          DO 100 J=3,39,4
0044    100   CLOSS(J)=(0.5*CLOSS(J-2)+CLOSS(J-1)+CLOSS(J)+
             1CLOSS(J+1)+0.5*CLOSS(J+2))*4.0
0045          DO 110 J=3,43,4
0046          CLOSS(J+1)=CLOSS(J)*3.14*(DIA(J)**2)
0047    110   CLOSS(J+2)=CLOSS(J)*0.523*(DIA(J)**3)
0048          TNUM=0.0
0049          TSURF=0.0
0050          TVOL=0.0
0051          DO 120 J=3,43,4
0052          TNUM=TNUM+CLOSS(J)
0053          TSURF=TSURF+CLOSS(J+1)
0054    120   TVOL=TVOL+CLOSS(J+2)
0055          WRITE(6,130)
0056    130   FORMAT(3X,'PORT NO.',3X,'OBS. DATA',3X,'CALC. DATA',
             13X,'MID PT. DIA',4X,'DNDLGD',5X,'DSDLGD',5X,'DVDLGD',/)
0057          I=0
0058          DO 150 J=3,39,4
0059          I=I+1
0060    150   WRITE(6,160) I,DAT(I),CDAT(I),DIA(J),CLOSS(J),
             1CLOSS(J+1),CLOSS(J+2)
0061    160   FORMAT(3X,I2,10X,F7.4,2X,F7.4,7X,F6.4,3(4X,1PE9.3))
0062          I=I+1
0063          WRITE(6,160)I,DAT(I),CDAT(I)
0064          GO TO (170,180) IBAT
0065    180   I=I+1
0066          WRITE(6,160) I,DAT(I),CDAT(I)
0067    170   TNUM=TNUM/4.0
0068          TSURF=TSURF/4.0
0069          TVOL=TVOL/4.0
0070          WRITE(6,190) TNUM,TSURF,TVOL
0071    190   FORMAT(3X,'TOTALS',25X,3(4X,1PE9.3))
        D     DO 195 J=3,39,4
        D     CLOSS(J+1)=CLOSS(J+1)/(TSURF*4.0)
        D     CLOSS(J+2)=CLOSS(J+2)/(TVOL*4.0)
        D195  CLOSS(J)=CLOSS(J)/(TNUM*4.0)
        D     WRITE(6,197)(CLOSS(J),J=3,39,4)
        D     WRITE(6,197)(CLOSS(J+1),J=3,39,4)
        D     WRITE(6,197) (CLOSS(J+2),J=3,39,4)
        D197  FORMAT(//3X,10(4X,F6.4),/)
0072          WRITE(5,200)
0073    200   FORMAT(3X,'ENTER 1 FOR ANOTHER CALC. 2 FOR STOP',/)
0074    205   READ(5,210) IFLAG
```

```
FORTRAN IV      V01C-03D          THU 16-AUG-79 11:48:35
CORE=08K. UIC=[100,103]                                          PAGE 003
                                                         ,LP:=DISKIA.ALL
0075  210   FORMAT(I1)
0076        IF (IFLAG .LT. 1 .OR. IFLAG .GT. 2) GO TO 205
0078        GO TO (230,220) IFLAG
0079  220   STOP
0080        END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:49:16          PAGE 001
CORE=08K, UIC=[100, 103]                                 ,LP:=DISKIA. ALL

      C      *DIFFTER. FTN
      C
      C      THIS SUBROUTINE USES A NONLINEAR ALGORITHM
      C      FOR SOLVING THE DFFUSION BATTERY EQNS.
      C
      C
0001         SUBROUTINE DIFFTER
0002         COMMON CALIB(12, 45), TDAT, KDAT, DAT(12), CDAT(12),
            1CLOSS(45), ITER, FLOW, DIA(45), X
0003         DIMENSION CNCEFF(45)
0004         DATA CNCEFF/45*1. 0/
0005         KMAX=20
0006         JMAX=45
0007         DO 10 J=1, JMAX
0008  10     CLOSS(J)=TDAT/JMAX
0009         ITER=0
0010         DO 40 I=1, KDAT
0011         CDAT(I)=0. 0
0012         DO 40 J=1, JMAX
0013  40     CDAT(I)=CDAT(I)+CALIB(I, J)*CLOSS(J)*CNCEFF(J)
0014  100    DO 20 K=1, KMAX
0015         DO 44 J=1, JMAX
0016         CORR=0. 0
0017         DO 71 I=1, KDAT
0018         IF (CDAT(I) .LT.  1. 0E-10) CDAT(I)=1. 0E-10
0020         IF (DAT(I) .LT.  1. 0E-10) DAT(I)=0. 0
0022  71     CORR=CORR+(DAT(I)/CDAT(I))*CALIB(I, J)*CNCEFF(J)
0023  44     CLOSS(J)=CLOSS(J)*CORR
0024         DO 70 I=1, KDAT
0025         CDAT(I)=0. 0
0026         DO 70 J=1, JMAX
0027  70     CDAT(I)=CDAT(I)+CLOSS(J)*CALIB(I, J)*CNCEFF(J)
0028  20     CONTINUE
0029         ITER=ITER+KMAX
0030         IF(ITER. GT. 100) GO TO 90
0032         CALL OBJCT
0033         IF(X .LT.  .005) GO TO 90
0035         KMAX=5
0036         GO TO 100
0037  90     CONTINUE
      D      WRITE(6, 630)
      D630   FORMAT(5X, 'DETAILED SOLN')
      D      WRITE(6, 620) CLOSS
      D620   FORMAT(10(3X, 1PE9. 3))
0038         RETURN
0039         END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:49:36          PAGE 001
CORE=08K, UIC=[100,103]                                 ,LP:=DISKIA.ALL

      C     *CHS.FTN
      C
      C     CHS.FTN SETS UP THE LOSS MATRIX
      C     FOR THE COLLIMATED HOLE STRUCTURE BATTERY
      C     FOR ANY SPECIFIED FLOW
      C
      C
      C
            SUBROUTINE CHS
            COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
           1CLOSS(45),ITER,FLOW,DIA(45),X
            DIMENSION ALEN(18),KTAP(11),P(18)
            DATA ALEN/2*0.3175,0.635,0.9525,1.27,1.905,
           12.54,1.27,10*2.54/
            DATA KTAP/1,2,3,4,5,6,7,9,11,14,18/
            DPI=0.00316228*.0001
            DPMULT=1.154781985
            WRITE(5,10)
      10    FORMAT(5X,'ENTER THE FLOW RATE(LPM) (F FORMAT)',/)
            READ(5,20) FLOW
      20    FORMAT(F13.0)
            FLOW=FLOW*16.6667
            DO 30 I=1,45
            D=(2.37E-11/DPI)*(1.0+(1.306E-5/DPI)*(1.246+
           10.42*EXP(-66616.0*DPI)))
            DO 40 J=1,18
            AMU=3.1459*ALEN(J)*D*9750./FLOW
            P(J)=0.819*EXP(-3.657*AMU)+0.097*EXP(-22.3*AMU)
           1+0.032*EXP(-57.0*AMU)+0.027*EXP(-123.0*AMU)
           2+0.025*EXP(-750.0*AMU)
      40    CONTINUE
            DO 50 J=2,12
            MUL=KTAP(J-1)
            CALIB(J-1,I)=1.0
            DO 60 KMUL=1,MUL
      60    CALIB(J-1,I)=CALIB(J-1,I)*P(KMUL)
      50    CONTINUE
            DPI=DPI*DPMULT
      30    CONTINUE
            DO 70 I=1,45
            P(1)=1.0-CALIB(1,I)
            DO 80 J=2,11
            P(J)=CALIB(J-1,I)-CALIB(J,I)
      80    CONTINUE
            P(12)=CALIB(11,I)
            DO 90 J=1,12
      90    CALIB(J,I)=P(J)
      70    CONTINUE
            RETURN
            END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:49:58          PAGE 001
CORE=08K, UIC=[100,103]                                    ,LP:=DISKIA.ALL

      C      *TSI.FTN
      C
      C      THIS SUBROUTINE COMPUTES THE LOSS MATRIX FOR
      C      TSI SCREEN BATTERY FOR 4.0 OR 6.0 LPM FLOW RATES.
      C
      C
0001          SUBROUTINE TSI
0002          COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
             1CLOSS(45),ITER,FLOW,DIA(45),X
0003   20     WRITE(5,10)
0004   10     FORMAT(5X,'IS THE FLOW RATE 4.0 OR 6.0 LPM?',2X,
             1'(F FORMAT)',)
0005          READ(5,30) FLOW
0006   30     FORMAT(F13.0)
0007          IF (FLOW .EQ. 4.0 .OR. FLOW .EQ. 6.0 ) GO TO 50
0009          GO TO 20
0010   50     IF(FLOW .EQ. 4.0 ) GO TO 60
0012          AK1=-1.1171981
0013          CK1=-3.067
0014          GO TO 70
0015   60     AK1=-1.14423
0016          CK1=-2.990605
0017   70     DO 90 J=1,45
0018          NSCRN=0
0019          SLOPE=-10.0**(AK1*ALOG10(DIA(J))+CK1)
0020          DO 80 I=2,11
0021          CALIB(I-1,J)=(10.0**(SLOPE*NSCRN))-(10.0**(SLOPE*(NSCRN+I-1)
             1))
0022   80     NSCRN=NSCRN+I-1
0023   90     CALIB(11,J)=10.0**(SLOPE*55.)
0024          RETURN
0025          END
```

```
                                                                 PAGE 001
FORTRAN IV      V01C-03D         THU 16-AUG-79 11:30:16    ,LP:=DISKIA.ALL
CORE=08K, UIC=[100,103]

      C       =WLS.FTN
      C
      C       THIS SUBROUTINE COMPUTES THE WLS FOR
      C       THE OBSERVED AND CALCULATED DATA FOR THE DIFFUSION BATTERY.
      C
      C
0001          SUBROUTINE OBJCT
0002          COMMON CALIB(12,45),TDAT,KDAT,DAT(12),CDAT(12),
             1CLOSS(45),ITER,FLOW,DIA(45),X
0003          X=0.0
0004          TCDAT=0.0
0005          DO 5 J=1,KDAT
0006    5     TCDAT=TCDAT+CDAT(J)
0007          DO 10J=1,KDAT
0008          AF=(CDAT(J)/TCDAT)-(DAT(J)/TDAT)
0009          IF(ABS(AF) .LT. .001 .AND. CDAT(J) .LT. .0001) GO TO 10
0011          X=X+(AF**2)/(DAT(J)/TDAT)
0012   10     CONTINUE
0013          RETURN
0014          END
```

APPENDIX G

COMPUTER PROGRAM FOR DB-CNC DATA REDUCTION USING SIMPLEX MINIMIZATION CURVE-FITTING PROCEDURE

(DIBWLS)

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:55:26          PAGE 001
CORE=08K, UIC=[100,103]                                   .LP:=DISWLS.ALL

        C       *DISWLS.FTN
        C
        C       DISWLS.FTN IS USED FOR DATA REDUCTION FOR A
        C       DIFFUSION BATTERY.  IN THIS PROGRAM THE DATA FROM CHS
        C       OR THE TSI SCREEN BATTERY CAN BE ANALYSED.  FOR THE TSI
        C       BATTERY THE FLOW RATES MUST BE 4.0 OR 6.0 LPM
        C       BUT FOR THE CHS BATTERY DATA WITH ANY FLOW RATE CAN BE
        C       ANALYSED.
        C
        C
        C
0001            COMMON /STEPIT/X(5),XMAX(5),XMIN(5),DELTAX(5),
               1DELMIN(5),MASK(5),CHISQ,BCHISQ,NF
0002            COMMON /ALOSS/CALIB(12,45),DAT(12),CLOSS(12),
               1CNUM1(45),CNUM2(45),CNUM(12),SD(11),TDAT,FLOW,CNCEFF(45)
               2,KDAT,CORFAC(12)
0003            DIMENSION TITTLE(10),ALPHA(5,5)
0004            DOUBLE PRECISION ALPHA
        C
        C
0005            CALL ASSIGN (6, 'LP:')
0006    360     WRITE(5,5)
0007    5       FORMAT(5X,'ENTER DATA DESCRIPTION (40 CHARACTERS)',/)
0008            READ(5,7) TITTLE
0009    7       FORMAT(10A4)
        C
        C       SET MASK  VALUES
        C
0010            WRITE(5,10)
0011    10      FORMAT(5X,'ONE OR TWO MODES?',/)
0012    12      READ (5,11) MODE
0013    11      FORMAT(I1)
0014            IF( MODE .EQ. 1 .OR. MODE .EQ. 2 ) GO TO 13
0016            GO TO 12
0017    13      MASK(1)=0
0018            MASK(2)=0
0019            IF(MODE .EQ. 2) GO TO 20
0021            DO 30 J=3,5
0022    30      MASK(J)=1
0023            GO TO 40
0024    20      DO 50 J=3,5
0025    50      MASK(J)=0
0026    40      IF (MODE .EQ. 2) GO TO 60
        C
        C       SET THE BOUNDS
        C
0028            WRITE(5,70)
0029    70      FORMAT(5X,'ENTER MINM. AND MAXM. FOR NMD1 AND SIGMA1',/)
0030            DO 80 J=1,2
0031    80      READ(5,81) XMIN(J),XMAX(J)
0032    81      FORMAT(2F12.0)
0033            XMIN(3)=0.1
0034            XMAX(3)=1.0
0035            XMIN(4)=1.1
```

```
FORTRAN IV      V01C-03D        THU 14-AUG-79 11:55:26          PAGE 002
CORE=08K, UIC=[100,103]                                      .LP:=GIDALS.ALL

0036          XMAX(4)=2.8
0037          XMIN(5)=0.0
0038          XMAX(5)=1.0
0039          GO TO 90
0040   60     WRITE(5,100)
0041   100    FORMAT(5X,'ENTER THE MINM. AND MAXM. VALUES FOR',/
             15X,'NMD1,SIGMA1,NMD2,SIGMA2,AND NUM. FRAC IN MODE ONE',/)
0042          DO 110 J=1,5
0043   110    READ(5,111) XMIN(J),XMAX(J)
0044   111    FORMAT(2F12.0)
       C
       C      SET THE APPROPRIATE LOSS MATRIX
       C
0045   90     WRITE(5,120)
0046   120    FORMAT (5X,'ENTER',/5X,'1 IF TBI SCREEN BATTERY WAS USED,'
             1/5X,'OR 2 IF CHB BATTERY WAS USED.',/)
0047   91     READ(5,121) IBAT
0048   121    FORMAT(I1)
0049          IF(IBAT .EQ. 1 .OR. IBAT .EQ. 2) GO TO 122
0051          GO TO 91
0052   122    IF (IBAT .EQ. 1) CALL TBI
0054          IF (IBAT .EQ. 2) CALL CHB
0056          DO 123 J=1,12
0057   123    DAT(J)=0.0
0058          IF(IBAT .EQ. 1) KDAT=10
0060          IF(IBAT .EQ. 2) KDAT=11
       C
       C      ENTER THE DATA FROM CHC
       C
0062   370    WRITE(5,130)
0063   130    FORMAT(5X,'ENTER THE DATA FROM CHC',/)
0064          READ(5,131) TDAT,(DAT(I),I=1,KDAT)
0065   131    FORMAT(12F13.0)
0066          KDAT=KDAT+1
0067          KDAT1=KDAT-1
0068          DATX=DAT(KDAT)
0069          DO 140 J=1,KDAT1
0070          KLAST=KDAT-J+1
0071          DAT(KLAST)=DAT(KLAST-1)-DAT(KLAST)
0072   140    DAT(KLAST)=AMAX1(DAT(KLAST),0.0)
0073          DAT(1)=AMAX1((TDAT-DAT(1)),0.0)
       D133   FORMAT(5X,'INPUT DATA',/13(X,1PE9.3),/3X,I2)
       C
       C      NORMALISE DATA
       C
0074          DO 150 J=1,KDAT
0075   150    DAT(J)=DAT(J)/TDAT
       D      WRITE(6,133) TDAT,DAT,KDAT
       C
       C      MINIMIZE
       C
0076   380    IF(MODE .EQ. 2) GO TO 160
0078          WRITE(5,170)
0079   170    FORMAT(5X,'ENTER THE ESTIMATES OF NMD1 AND SIGMA1',/)
```

```
            READ(5,171) X(1),X(2)
      171   FORMAT(2F13.0)
            X(3)=.1
            X(4)=2.0
            X(5)=1.0
            GO TO 180
      160   WRITE(5,190)
      190   FORMAT(5X,'ENTER THE ESTIMATES OF MMD1,SIGMA1,MMD2,',
           1/5X,'SIGMA2 AND NUM. FRAC IN MODE ONE',/)
            READ(5,191) X
      191   FORMAT(5F13.0)
      C
      C     SET THE PARAMETERS FOR MINIMIZATION
      C
      180   DO 400 J=1,5
            DELTAX(J)=DELTAX(J)*X(J)
      400   DELMIN(J)=DELMIN(J)*X(J)
            CALL STEPIT
            FCHISQ=CHISQ
      C
      C     COMPUTE THE FINAL DISTRIBUTION
      C
            CALL CNUMS(CNUM1,X(1),X(2),X(5))
            FRAC2=1.0-X(5)
            CALL CNUMS(CNUM2,X(3),X(4),FRAC2)
            DO 200 J=1,45
      200   CNUM1(J)=CNUM1(J)+CNUM2(J)
            TCNC=0.0
            THORM=0.0
            DO 205 J=1,45
            THORM=THORM+CNUM1(J)
      205   TCNC=TCNC+CNUM1(J)*CNCEFF(J)
            TCNC=(TDAT*THORM)/TCNC
            DO 210 J=1,KDAT
            CORFAC(J)=DAT(J)/CLOSS(J)
      210   CONTINUE
            DO 220 I=1,45
            CNUM2(I)=0.0
            DO 220 J=1,KDAT
      220   CNUM2(I)=CNUM2(I)+CNUM1(I)*CORFAC(J)*CALIB(J,I)
            CMTOT=0.0
            DO 230 J=1,45
      230   CMTOT=CMTOT+CNUM2(J)
            DO 240 J=1,45
      240   CNUM2(J)=CNUM2(J)/CMTOT
            I=1
            DO 250 J=3,43,4
            CNUM(I)=(0.5*CNUM2(J-2)+CNUM2(J-1)+CNUM2(J)+CNUM2(J+1)
           1+0.5*CNUM2(J+2))*TCNC
      250   I=I+1
            WRITE(5,630)
      630   FORMAT(5X,'ENTER 1 IF YOU WANT ERROR ANALYSIS',/
           15X,'OR 2 IF YOU DO NOT',/)
            READ (5,341) IFLAG
```

```
FORTRAN IV      V01C-03D          THU 16-AUG-79 11:55:26           PAGE 004
CORE=08K, UIC=[100,103]                                      ,LP:=DISMLS.ALL

0125            IF (IFLAG .EQ. 1) GO TO 640
0127            GO TO 650
0128      640   WRITE(5,660)
0129      660   FORMAT(5X,'ENTER S.E. PROPORTIONALITY CONSTANT',/
               15X,'(0.075 FOR LABORATORY AEROSOL OR',/
               25X,' 0.15 FOR ATMOSPHERIC AEROSOL)',/)
0130            READ(5,131) CSIG
0131            CALL ERRMAT(ALPHA,MODE)
0132            DO 670 I=1,5
0133      670   ALPHA(I,I)=SQRT(ALPHA(I,I))*CSIG
          C
          C     OUTPUT
          C
0134      650   WRITE(6,560) TITTLE
0135      560   FORMAT(//5X,10A4,/)
0136            WRITE(6,260)BCHISQ,FCHISQ,NF,MODE
0137      260   FORMAT(5X,'BEGIN OBJCT.',21X,'FINAL OBJCT.',/5X,'FCT.  VALUE  =',
               11PE10.3,10X,'FCT.  VALUE  =',1PE10.3,
               1//5X,0PI3,' COMPUTATIONS',
               217X,'MODE(S)=',I1)
0138            IF (MODE .EQ. 2) GO TO 270
0140            WRITE(6,280) X(1),X(2)
0141      280   FORMAT(/5X,'NMD1=',F7.4,5X,'SIGMA1=',F4.2,/)
0142            IF (IFLAG .EQ. 1) WRITE(6,680) ALPHA(1,1),ALPHA(2,2)
0144      680   FORMAT(/5X,'STANDARD ERROR ESTIMATES',/
               15X,'NMD1=',1PE7.1,3X,'SIGMA1=',1PE7.1)
0145            GO TO 290
0146      270   WRITE(6,300)X
0147      300   FORMAT(/5X,'NMD1=',F7.4,5X,'SIGMA1=',F4.2,/5X,'NMD2=',
               1F7.4,5X,'SIGMA2=',F4.2,/5X,'FRAC1=',F4.2,/)
0148            IF (IFLAG .EQ. 1) WRITE(6,690) ALPHA(1,1),ALPHA(2,2),
               1ALPHA(3,3),ALPHA(4,4),ALPHA(5,5)
0150      690   FORMAT(/5X,'STANDARD ERROR ESTIMATES',/
               15X,'NMD1=',1PE7.1,1X,'SIGMA1=',1PE7.1,
               21X,'NMD2=',1PE7.1,1X,'SIGMA2=',1PE7.1,/
               35X,'NUMBER FRACTION IN FIRST MODE=',1PE7.1)
0151      290   WRITE(6,310) SD(1)
0152      310   FORMAT(//22X,'INPUT DATA',20X,'RESULTS',/
               15X,'SAT PORT NO.',5X,'CMC DATA',5X,'HSTGRM SDS.',
               27X,'NUMBER/CC',/38X,F6.4)
0153            DO 315 J=1,KDAT
0154            CNUM2(J)=0.0
0155            DO 317 J1=J,KDAT
0156      317   CNUM2(J)=CNUM2(J)+DAT(J1)
0157      315   CONTINUE
0158            DO 319 J=1,KDAT
0159      319   CNUM2(J)=CNUM2(J)*TDAT
0160            DO 320 J=1,10
0161      320   WRITE(6,330) J,CNUM2(J),CNUM(J),SD(J+1)
0162            J1=11
0163            IF( KDAT .EQ. 12) WRITE(6,330) J1,CNUM2(J1)
0165            WRITE(6,330) KDAT,CNUM2(KDAT)
0166      330   FORMAT(7X,I2,13X,F7.3,20X,1PE10.3,/38X,0PF6.4)
          D     DO 333 J=1,10
```

```
FORTRAN IV      V01C-03D         THU 16-AUG-79 11:55:26          PAGE 005
CORE=08K, UIC=[100,103]                                        ,LP:=DISHLS.ALL

       D333  CNUM1(J)=CNUM(J)/TCNC
       D     WRITE(6,335)(CNUM1(J),J=1,10)
       D335  FORMAT(//5X,10(4X,F6.4))
0167         WRITE(5,340)
0168   340   FORMAT(5X,'ENTER 1 IF YOU WANT TO STOP.'
            1/5X,'2 IF YOU WANT TO START FROM THE BEGINING.'
            2/5X,'3 IF YOU WANT TO ENTER NEW DATA FROM CNC.'
            3/5X,'OR 4 IF YOU WANT TO ENTER NEW ESTIMATES ONLY.'/)
0169         DELTAX(1)=0.5
0170         DELTAX(3)=0.5
0171         DELTAX(2)=0.25
0172         DELTAX(4)=0.25
0173         DELTAX(5)=0.15
0174         DELMIN(1)=0.1
0175         DELMIN(3)=0.1
0176         DELMIN(2)=0.05
0177         DELMIN(4)=0.05
0178         DELMIN(5)=0.05
0179   347   READ(5,341) IFLAG
0180         IF(IFLAG .LT. 1 .OR. IFLAG .GT. 4) GO TO 347
0182   341   FORMAT(I1)
0183         GO TO (350,360,370,380) IFLAG
0184   350   STOP
0185         END
```

```
                                                                PAGE 001
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:56:42     .LP:=DISMLS.ALL
CORE=08K, UIC=[100,103]

       C        *XLOSS.FTN
       C
       C        THIS SUBROUTINE CALCULATES THE THEORETICAL LOSS
       C        OF AN AEROSOL DISTRIBUTION WHEN PASSED THROUGH A
       C        DIFFUSION BATTERY.
       C
       C
0001            SUBROUTINE XLOSS
0002            COMMON/ALOSS/CALIB(12,45),DAT(12),CLOSS(12),
               1CNUM1(45),CNUM2(45),CNUM(12),SD(11),TDAT,FLOW,CNCEFF(45)
               2,KDAT,CORFAC(12)
0003            COMMON/STEPIT/X(5),XMAX(5),XMIN(5),DELTAX(5),
               1DELMIN(5),MASK(5),CHISQ,SCHISQ,NF
0004            CALL CNUMB(CNUM1,X(1),X(2),X(5))
0005            FRAC2=1.0-X(5)
0006            CALL CNUMB(CNUM2,X(3),X(4),FRAC2)
0007            DO 10 J=1,45
0008     10     CNUM1(J)=CNUM1(J)+CNUM2(J)
0009            DO 20 J=1,KDAT
0010            CLOSS(J)=0.0
0011            DO 20 I=1,45
0012     20     CLOSS(J)=CLOSS(J)+CALIB(J,I)*CNUM1(I)*CNCEFF(I)
0013            CLTOT=0.0
0014            DO 30 J=1,KDAT
0015     30     CLTOT=CLTOT+CLOSS(J)
0016            DO 40 J=1,KDAT
0017     40     CLOSS(J)=CLOSS(J)/CLTOT
0018            RETURN
0019            END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:57:01           PAGE 001
CORE=08K, UIC=[100,103]                                       ,LP:=DIBWLS.ALL

      C      ERRMAT.FTN CALCULATES THE CURVATURE MATRIX FOR
      C      FOR THE OBJECTIVE FUNCTION
      C
0001         SUBROUTINE ERRMAT(ALPHA,MODE)
0002         COMMON /STEPIT/X(5),XMAX(5),XMIN(5),DELTAX(5),
            1DELMIN(5),MASK(5),CHISQ,BCHISQ,NF
0003         COMMON /ALOSS/CALIB(12,45),DAT(12),CLOSS(12),
            1CNUM1(45),CNUM2(45),CNUM(12),BD(11),TDAT,FLOW,CNCEFF(45)
            2,KDAT,CORFAC(12)
      C
0004         DIMENSION ALPHA(5,5),DELTA(5)
0005         DOUBLE PRECISION ALPHA
0006         DELTA(1)=0.7783*X(1)
0007         DELTA(2)=10.**(ALOG10(X(2))*1.2) -X(2)
0008         IF (DELTA(2) .LT. .1) DELTA(2)=.1
0010         DELTA(3)=0.7783*X(3)
0011         DELTA(4)=10.**(ALOG10(X(4))*1.2)-X(4)
0012         IF (DELTA(4) .LT. .1) DELTA(4)=.1
0014         DELTA(5)=X(5)*(1.0-X(5))*1.2
0015         IF (DELTA(5) .GT. .2 ) DELTA(5)=.2
0017         DO 100 J=1,5
0018         XJ=X(J)+DELTA(J)
0019         IF (XMAX(J) -XJ) 120,110,110
0020  120    DELTA(J)=XMAX(J)-X(J)
0021  110    XJ=X(J)-DELTA(J)
0022         IF( XMIN(J)-XJ) 100,100,130
0023  130    DELTA(J)=X(J)-XMIN(J)
0024  100    CONTINUE
0025         IF (MODE .EQ. 1) NV=2
0027         IF(MODE .EQ. 2) NV=5
0029         FCHISQ=CHISQ
0030         DO 5 J=1,NV
0031         DO 5 K=1,NV
0032  5      ALPHA(J,K)=0.0
0033         DO 10 J=1,NV
0034         XJ=X(J)
0035         X(J)=X(J)+DELTA(J)
0036         CALL OBJCT (NV)
0037         CHISQ2=CHISQ
0038         ALPHA(J,J)=CHISQ2-2.0*FCHISQ
0039         DO 20 K=1,NV
0040         IF(K-J) 30,20,40
0041  30     ALPHA(K,J)=(ALPHA(K,J)-CHISQ2)/2.0
0042         ALPHA(J,K)=ALPHA(K,J)
0043         GO TO 20
0044  40     ALPHA(J,K)=FCHISQ-CHISQ2
0045         XK=X(K)
0046         X(K)=X(K)+DELTA(K)
0047         CALL OBJCT(NV)
0048         CHISQ3=CHISQ
0049         ALPHA(J,K)=ALPHA(J,K)+CHISQ3
0050         X(K)=XK
0051  20     CONTINUE
0052         X(J)=XJ-DELTA(J)
```

```
                                              PAGE 002
FORTRAN IV      V01C-03D      THU 16-AUG-79 11:37:01      ,LP:=DIBHLS.ALL
CORE=08K, UIC=[100,103]

0053            CALL OBJCT (NV)
0054            CHISQ3=CHISQ
0055            X(J)=XJ
0056            ALPHA(J,J)=(ALPHA(J,J)+CHISQ3)/2.0
0057     10     CONTINUE
0058            DO 50 J=1,NV
0059            DO 50 K=1,NV
0060            ALPHA(J,K)=ALPHA(J,K)/(DELTA(J)*DELTA(K))
0061     50     CONTINUE
0062            CALL MATINV (ALPHA, NV,DET)
0063     80     RETURN
0064            END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:57:31          PAGE 001
CORE=08K. UIC=[100.103]                                   .LP:=OIBWLS.ALL

        C
        C       SUBROUTINE MATINV INVERTIS A SYMMETRIC MATRIX AND CALCULATES
        C       ITS DETERMINANT
        C
        C       FROM BEVINGTON BASED ON IBM SCIENTIFIC SUBROUTINE PACKAGE
        C
        C
0001            SUBROUTINE MATINV(ARRAY.NORDER.DET)
0002            DOUBLE PRECISION ARRAY. AMAX. SAVE
0003            DIMENSION ARRAY(5.5). IK(5). JK(5)
        D       WRITE(6.250) ARRAY
        D250    FORMAT (5(3X.1PE10.3))
0004    10      DET=1. 0
0005    11      DO 100 K=1.NORDER
        C
        C       FIND THE LARGEST ELEMENT ARRAY(I.J) IN REST OF MATRIX
        C
0006            AMAX=0. 0
0007    21      DO 30 I=K.NORDER
0008            DO 30 J=K.NORDER
0009    23      IF (DABS(AMAX) -DABS( ARRAY (I.J))) 24.24.30
0010    24      AMAX=ARRAY(I.J)
0011            IK(K)=I
0012            JK(K)=J
        D       WRITE(6.150) ARRAY(I.J).AMAX.K.I.J
        D150    FORMAT(3X.'30'.2(3X.1PE10.3).3(5X.OPI3))
0013    30      CONTINUE
        C
        C       INTERCHANGE ROWS AND COLUMNS TO PUT AMAX IN ARRRAY(K.K)
        C
0014    31      IF (AMAX) 41.32.41
0015    32      DET=0. 0
0016            GO TO 140
0017    41      I=IK(K)
0018            IF (I-K) 21.51.43
0019    43      DO 50 J=1.NORDER
0020            SAVE= ARRAY(K.J)
0021            ARRAY(K.J)= ARRAY(I.J)
        D       WRITE (6.220) ARRAY(K.J).SAVE.K.I.J
0022    50      ARRAY(I.J)=-SAVE
        D220    FORMAT(3X.'51'.2(3X.1PE10.3).3(5X.OPI3))
0023    51      J=JK(K)
0024            IF (J-K) 21.61.53
0025    53      DO 60 I=1.NORDER
0026            SAVE=ARRAY(I.K)
0027            ARRAY(I.K)=ARRAY(I.J)
        D       WRITE(6.230) ARRAY(I.K).SAVE.K.I.J
0028    60      ARRAY(I.J)=-SAVE
        D230    FORMAT(3X.'61'.2(3X.1PE10.3).3(5X.OPI3))
        C
        C       ACCUMULATE ELEMENTS OF INVERSE MATRIX
        C
0029    61      DO 70 I=1.NORDER
0030            IF (I-K) 63.70.63
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:57:31          PAGE 002
CORE=08K, UIC=[100,103]                                     ,LP:=DIBHLS.ALL

0031   63     ARRAY(I,K)=-ARRAY(I,K)/AMAX
       D      WRITE(6,160) ARRAY(I,K),AMAX,K,I
       D160   FORMAT(5X,'70',2(3X,1PE10.3),2(5X,0PI3))
0032   70     CONTINUE
0033   71     DO 80 I=1,NORDER
0034          DO 80 J=1,NORDER
0035          IF (I-K) 74,80,74
0036   74     IF (J-K) 75,80,75
0037   75     ARRAY(I,J)=ARRAY(I,J)+ARRAY(I,K)*ARRAY(K,J)
       D      WRITE(6,170) ARRAY(I,J),ARRAY(I,K),ARRAY(K,J),K,I,J
       D170   FORMAT(5X,'80',3(3X,1PE10.3),3(5X,0PI3))
0038   80     CONTINUE
0039   81     DO 90 J=1,NORDER
0040          IF (J-K) 83,90,83
0041   83     ARRAY(K,J)=ARRAY(K,J)/AMAX
       D      WRITE(6,180) ARRAY(K,J),AMAX,K,J
       D180   FORMAT(5X,'90',2(3X,1PE10.3),2(5X,0PI3))
0042   90     CONTINUE
0043          ARRAY(K,K)=1.0/AMAX
       D      WRITE(6,190) ARRAY(K,K),AMAX,K
       D190   FORMAT(4X,'100',2(3X,1PE10.3),5X,0PI3)
0044   100    DET=DET*AMAX
       C
       C      RESTORE ORDERING OF MATRIX
       C
0045   101    DO 130 L=1,NORDER
0046          K=NORDER-L+1
0047          J=IK(K)
0048          IF (J-K) 111,111,105
0049   105    DO 110 I=1,NORDER
0050          SAVE=ARRAY(I,K)
0051          ARRAY(I,K)=-ARRAY(I,J)
       D      WRITE(6,200)ARRAY(I,J),SAVE,K,I,J
       D200   FORMAT(5X,'110',2(3X,1PE10.3),3(5X,0PI3))
0052   110    ARRAY(I,J)=SAVE
0053   111    I=JK(K)
0054          IF(I-K) 130,130,113
0055   113    DO 120 J=1,NORDER
0056          SAVE=ARRAY(K,J)
0057          ARRAY(K,J)=-ARRAY(I,J)
       D      WRITE(6,210) ARRAY(I,J),SAVE,K,I,J
       D210   FORMAT(4X,'120',2(3X,1PE10.3),3(5X,0PI3))
0058   120    ARRAY(I,J)=SAVE
0059   130    CONTINUE
       D      WRITE(6,250) ARRAY
0060   140    RETURN
0061          END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:38:00          PAGE 001
CORE=08K, UIC=[100,103]                                  .LP:=DISHLS.ALL

0001          SUBROUTINE STEPIT
        C
        C  MINIMIZES A PIECEWISE CONTINUOUS FUNCTION SUBJECT TO ARBITRARY
        C  CONSTRAINTS OF INEQUALITY.
        C  J. A. NELDER AND R. MEAD, THE COMPUTER JOURNAL 7 (1965) 308
        C
        C
        C
0002          COMMON/STEPIT/X(5),XMAX(5),XMIN(5),DELTAX(5),
             1DELMIN(5),MASK(5),CHISQ,BCHIS[illegible] NF
0003          DIMENSION CHI(6),Z(6,5),ZBAR(5),ZSTAR(5)
        C
0004          NFLAT=1
0005          CHISQ=0.0
0006          NV=5
0007          NFMAX=500
0008       40 HUGE=0.5E 38
0009          ALPHA=1.0
0010          BETA=0.5
0011          GAMMA=2.0
        C
0012       50 DO 130 J=1,NV
0013          IF(MASK(J))130,60,130
0014       60 IF((X(J)+DELTAX(J))-X(J))100,70,100
0015       70 IF(X(J))90,80,90
0016       80 DELTAX(J)=0.01
0017          GO TO 100
0018       90 DELTAX(J)=0.01*X(J)
0019      100 IF(XMAX(J)-XMIN(J))110,110,120
0020      110 XMAX(J)=HUGE
0021          XMIN(J)=-HUGE
0022      120 X(J)=AMAX1(XMIN(J),AMIN1(XMAX(J),X(J)))
0023      130 CONTINUE
        C
        C
        C  CALCULATE INITIAL P(I) AND Y(I).
        C
0024      220 NVA=0
0025          NF=0
0026          J=1
0027      230 IF(MASK(J))270,240,270
0028      240 NVA=NVA+1
0029          DO 250 K=1,NV
0030      250 Z(NVA,K)=X(K)
0031          Z(NVA,J)=Z(NVA,J)+DELTAX(J)
0032          XS=X(J)
0033          X(J)=Z(NVA,J)
0034          CALL OBJCT(NV)
0035      260 X(J)=XS
0036          CHI(NVA)=CHISQ
0037      270 J=J+1
0038          IF(J-NV)230,230,280
0039      280 IF(NVA)290,290,310
0040      290 DO 300 J=1,NV
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:58:00          PAGE 002
CORE=08K, UIC=[100,103]                                     ,LP:=DISMLB.ALL

      300 MASK(J)=0
          GO TO 30
      310 NVP=NVA+1
          DO 320 J=1,NV
      320 Z(NVP,J)=X(J)
C
C
          CALL OBJCT(NV)
      390 CHI(NVP)=CHISQ
          BCHISQ=CHISQ
      440 JL=NVP
C
C  DETERMINE H AND L.
C
      450 JH=JL
          DO 490 J=1,NVP
          IF(CHI(J)-CHI(JH))470,470,460
      460 JH=J
      470 IF(CHI(J)-CHI(JL))480,490,490
      480 JL=J
      490 CONTINUE
          IF(JH-JL)560,500,560
      500 IF(NFLAT)540,540,1200
      540 JH=1
          IF(JL-1)560,550,560
      550 JH=2
C
C  CALCULATE PBAR.
C
      560 DO 600 J=1,NV
          IF(MASK(J))600,570,600
      570 ZBAR(J)=0.0
          DO 590 K=1,NVP
          IF(K-JH)580,590,580
      580 ZBAR(J)=ZBAR(J)+Z(K,J)
      590 CONTINUE
          ZBAR(J)=ZBAR(J)/FLOAT(NVA)
      600 CONTINUE
C
C  ATTEMPT A REFLECTION.
C  FORM P* .
C
          DO 620 J=1,NV
          IF(MASK(J))620,610,620
      610 X(J)=(1.0+ALPHA)*ZBAR(J)-ALPHA*Z(JH,J)
      620 ZSTAR(J)=X(J)
          CALL OBJCT(NV)
      630 CHISTR=CHISQ
          IF(CHISQ-CHI(JL))690,730,730
C
C  THE REFLECTION SUCCEEDED.  ATTEMPT AN EXPANSION
C  FORM P** .
C
      690 DO 710 J=1,NV
```

```
FORTRAN IV      V01C-03B        THU 16-AUG-79 11:58:00          PAGE 003
CORE=08K, UIC=[100,103]                                      .LP:=DISHLS.ALL

0079           IF(MASK(J))710, 700, 710
0080       700 X(J)=GAMMA*X(J)+(1. 0-GAMMA)*ZBAR(J)
0081       710 CONTINUE
0082           CALL OBJCT(NV)
0083       720 IF(CHISQ-CHISTR)1100, 790, 790
        C
        C  THE REFLECTION FAILED.
        C
0084       730 DO 750 J=1, NVP
0085           IF(J-JH)740, 750, 740
0086       740 IF(CHISQ-CHI(J))790, 750, 750
0087       750 CONTINUE
0088           IF(CHISQ-CHI(JH))810, 830, 830
        C
        C
        C  REPLACE P(JH) BY P*
        C
0089       790 DO 800 J=1, NV
0090       800 Z(JH, J)=ZSTAR(J)
0091           CHI(JH)=CHISTR
0092           GO TO 1120
        C
0093       810 DO 820 J=1, NV
0094       820 Z(JH, J)=X(J)
0095           CHI(JH)=CHISQ
        C
        C  ATTEMPT A CONTRACTION.
        C  FORM P** .
        C
0096       830 DO 850 J=1, NV
0097           IF(MASK(J))850, 840, 850
0098       840 X(J)=BETA*Z(JH, J)+(1. 0-BETA)*ZBAR(J)
0099       850 CONTINUE
0100           CALL OBJCT(NV)
0101       860 IF(CHISQ-CHI(JH))1100, 960, 960
        C
        C  THE CONTRACTION FAILED.
        C  REPLACE ALL P(I) BY (P(I)+P(JL))/2.0 , GUARDING THE ROUNDING.
        C
0102       960 JS=JL
0103           DO 1060 J=1, NVP
0104           IF(J-JL)970, 1060, 970
0105       970 DO 1020 K=1, NV
0106           IF(MASK(K))1020, 980, 1020
0107       980 IF(Z(J, K)-Z(JL, K))990, 1000, 990
0108       990 X(K)=(Z(J, K)+Z(JL, K))/2. 0
0109           IF(X(K)-Z(J, K))1010, 1000, 1010
0110      1000 X(K)=Z(JL, K)
0111      1010 Z(J, K)=X(K)
0112      1020 CONTINUE
0113           CALL OBJCT(NV)
0114      1030 IF(CHISQ-CHI(JS))1040, 1050, 1050
0115      1040 JS=J
0116      1050 CHI(J)=CHISQ
```

```
FORTRAN IV      V01C-03D          THU 16-AUG-79 11:58:00              PAGE 004
CORE=08K, UIC=[100,103]                                           ,LP:=DISMLS.ALL

0117    1060 CONTINUE
0118          JL=JH
0119          GO TO 1120
        C
        C
        C  REPLACE P(JH) BY P* OR P** .
        C
0120    1100 DO 1110 J=1,NV
0121    1110 Z(JH,J)=X(J)
0122          CHI(JH)=CHISQ
        C
        C  TEST FOR CONVERGENCE.
        C
0123    1120 DO 1150 J=1,NV
0124          IF(MASK(J))1150,1130,1150
0125    1130 ZMAX=Z(1,J)
0126          ZMIN=Z(1,J)
0127          DO 1140 K=2,NVP
0128          ZMAX=AMAX1(ZMAX,Z(K,J))
0129    1140 ZMIN=AMIN1(ZMIN,Z(K,J))
0130          IF((ZMAX-ZMIN)-ABS(DELMIN(J)))1150,1150,1260
0131    1150 CONTINUE
        C
0132    1180  IF(CHI(JH)-CHI(JL))1190,1200,1200
0133    1190  JL=JH
0134    1200 DO 1210 J=1,NV
0135    1210 X(J)=Z(JL,J)
0136          CALL OBJCT(NV)
0137          FCHISQ=CHISQ
0138    1250 RETURN
        C
0139    1260  CONTINUE
0140    1270 IF(NF-NFMAX)450,450,1180
0141          END
```

```
FORTRAN IV      V01C-03B        THU 16-AUG-79 11:39:00         PAGE 001
CORE=08K, UIC=[100,103]                                   ,LP:=SIGMLS.ALL

      C       THIS SUBROUTINE CALCULATES THE NUMBER IN THE 45 DIVISIONS
      C
0001          SUBROUTINE CNUMB(CNUM,ANMD,SIGMA,FRAC)
0002          DIMENSION CNUM(45)
0003          SD1=0.00294273
0004          DPMULT=1.194781985
0005          XANMD=ALOG(ANMD)
0006          XSIGMA=ALOG(SIGMA)
0007          DO 10 J=1,45
0008          A=(ALOG(SD1)-XANMD)/XSIGMA
0009          B=(ALOG(SD1*DPMULT)-XANMD)/XSIGMA
0010          SD1=SD1*DPMULT
0011    10    CNUM(J)=APROX(A,B)*FRAC
0012          RETURN
0013          END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:39:10          PAGE 001
CORE=08K, UIC=[100,103]                                    ,LP:=SIGNALS.ALL

      C       THIS IS AN APPROXIMATION FOR THE CUMULATIVE
      C       NORMAL DISTRIBUTION FUNCTION. REF. ABRAMOWITZ
      C       AND STEGUN: MATHEMATICAL HANDBOOK.
              FUNCTION APROX(A,B)
              X1=ABS(A)
              X2=ABS(B)
              IF(X1) 10,20,10
      10       PX1=P(X1)
              GO TO 30
      20       PX1=0.5
      30       IF(X2) 35,40,35
      35       PX2=P(X2)
              GO TO 45
      40       PX2=0.5
      45       SIN=A*B
              IF(SIN)60,50,50
      50       APROX=ABS(PX2-PX1)
              GO TO 70
      60       APROX=1.0-(PX2-PX1)
      70       RETURN
              END
```

```
FORTRAN IV      V01C-03D          THU 16-AUG-79 11:59:21               PAGE 001
CORE=08K, UIC=[100,103]                                            ,LP:=DIBWLS.ALL

      C        THIS FUNCTION IS USED WITH APROX
0001           FUNCTION P(X)
0002           DIMENSION B(6)
0003           DATA B/0.319381530,-0.356563782,1.781477937,-1.821255978,
              11.330274429,0.2316419/
0004           T=1.0/(1.0+B(6)*X)
0005           P=Z(X)*(B(1)*T+B(2)*T**2+B(3)*T**3+B(4)*T**4+B(5)*T**5)
0006           RETURN
0007           END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:39:29          PAGE 001
CORE=08K, UIC=[100,103]                                     .LP:=DISHLS.ALL

      C     THIS FUNCTION IS USED WITH APPROX
            FUNCTION Z(X)
            PI=3.1415927
            S2PI=SQRT(2.0*PI)
            ARG=X**2/2.0
            IF(ARG-87.0) 10,20,20
      10     A=EXP(ARG)
            Z=1.0/(S2PI*A)
            RETURN
      20     Z=0.0
            RETURN
            END
```

```
FORTRAN IV      V01C-03D           THU 14-AUG-79 11:59:39
CORE=08K. UIC=[100.103]                                              PAGE 001
                                                             .LP:=DISHLS.ALL
        C       *TSI.FTN
        C
        C       THIS SUBROUTINE COMPUTES THE LOSS MATRIX FOR
        C       TSI SCREEN BATTERY FOR 4.0 OR 6.0 LPM FLOW RATES.
        C
        C
0001            SUBROUTINE TSI
0002            COMMON /ALOSS/CALIB(12,45),DAT(12),CLOSS(12),
               1CNUM1(45),CNUM2(45),CNUM(12),SD(11),TDAT,FLOW,CNCEFF(45),
               2KDAT,CORFAC(12)
0003            DPI=0.00316228
0004            DPMULT=1.154781983
0005     20     WRITE(5,10)
0006     10     FORMAT(3X,'IS THE FLOW RATE 4.0 OR 6.0 LPM?',2X,
               1'(F FORMAT)',)
0007            READ(5,30) FLOW
0008     30     FORMAT(F10.0)
0009            IF (FLOW .EQ. 4.0 .OR. FLOW .EQ. 6.0 ) GO TO 50
0011            GO TO 20
0012     50     IF(FLOW .EQ. 4.0 ) GO TO 60
0014            AK1=-1.1171981
0015            CK1=-3.067
0016            GO TO 70
0017     60     AK1=-1.14423
0018            CK1=-2.990605
0019     70     DO 80 J=1,45
0020            NSCRN=0
0021            SLOPE=-10.0**(AK1*ALOG10(DPI)+CK1)
0022            DO 90 I=2,11
0023            CALIB(I-1,J)=(10.0**(SLOPE*NSCRN))-(10.0**(SLOPE*(NSCRN+I-1)
               1))
0024     90     NSCRN=NSCRN+I-1
0025            CALIB(11,J)=10.0**(SLOPE*55.)
         D      WRITE(6,100)(CALIB(I1,J),I1=1,11),DPI
0026     80     DPI=DPI*DPMULT
         D100   FORMAT(12(3X,F6.4))
0027            RETURN
0028            END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 11:39:56          PAGE 001
CORE=08K, UIC=[100,103]                                     ,LP:=DISMLS.ALL

      C       *CHS.FTN
      C
      C       CHS.FTN SETS UP THE LOSS MATRIX
      C       FOR THE COLLIMATED HOLE STRUCTURE BATTERY
      C       FOR ANY SPECIFIED FLOW
      C
      C
      C
0001          SUBROUTINE CHS
0002          COMMON/ALOSS/CALIB(12,45),DAT(12),CLOSS(12),
             1CNUM1(45),CNUM2(45),CNUM(12),BD(11),TDAT,FLOW,CNCEFF(45),
             2XDAT,CORFAC(12)
0003          DIMENSION ALEN(18),KTAP(11),P(18)
0004          DATA ALEN/2*0.3175,0.635,0.9525,1.27,1.905,
             12.54,1.27,10*2.54/
0005          DATA KTAP/1,2,3,4,5,6,7,9,11,14,18/
0006          DPI=0.00316228*.0001
0007          DPMULT=1.154781985
0008          WRITE(5,10)
0009    10    FORMAT(3X,'ENTER THE FLOW RATE(LPM) (F FORMAT)',/)
0010          READ(5,20) FLOW
0011    20    FORMAT(F13.0)
0012          FLOW=FLOW*16.6667
0013          DO 30 I=1,45
0014          D=(2.37E-11/DPI)*(1.0+(1.306E-5/DPI)*(1.246+
             10.42*EXP(-66616.0*DPI)))
0015          DO 40 J=1,18
0016          AMU=3.1459*ALEN(J)*D*9750./FLOW
0017          P(J)=0.819*EXP(-3.657*AMU)+0.097*EXP(-22.3*AMU)
             1+0.032*EXP(-57.0*AMU)+0.027*EXP(-123.0*AMU)
             2+0.025*EXP(-750.0*AMU)
0018    40    CONTINUE
0019          DO 50 J=2,12
0020          MUL=KTAP(J-1)
0021          CALIB(J-1,I)=1.0
0022          DO 60 KMUL=1,MUL
0023    60    CALIB(J-1,I)=CALIB(J-1,I)*P(KMUL)
0024    50    CONTINUE
0025          DPI=DPI*DPMULT
0026    30    CONTINUE
0027          DO 70 I=1,45
0028          P(1)=1.0-CALIB(1,I)
0029          DO 80 J=2,11
0030          P(J)=CALIB(J-1,I)-CAL[illegible](J,I)
0031    80    CONTINUE
0032          P(12)=CALIB(11,I)
0033          DO 90 J=1,12
0034    90    CALIB(J,I)=P(J)
0035    70    CONTINUE
      D100    FORMAT(12(3X,F6.4))
      D       WRITE(6,100) ((CALIB(I,J),I=1,12),J=1,45)
0036          RETURN
0037          END
```

```
FORTRAN IV      V01C-03D        THU 16-AUG-79 12:00:18        PAGE 001
CORE=08K, UIC=[100,102]                               ,LP:=[illegible].ALL

        C       *OBJECT FTN
        C
        C       THIS SUBROUTINE CALCULATES THE FUNCTION TO BE MINIMISED.
        C
0001            SUBROUTINE OBJCT(NV)
0002            COMMON/ALOSS/CALIB(12,45),DAT(12),CLOSS(12),
               1CNUM1(45),CNUM2(45),CNUM(12),SD(11),TDAT,FLOW,CNCEFF(45),
               2NDAT,CORFAC(12)
0003            COMMON/STEPIT/X(5),XMAX(5),XMIN(5),DELTAX(5),DELMIN(5),
               1MASK(5),CHISQ,SCHISQ,NF
0004            CHISQ=0.0
0005            DO 10 J=1,NV
0006            IF(MASK(J))10,20,10
0007    20      IF(X(J)-XMAX(J))30,30,40
0008    30      IF(X(J)-XMIN(J))40,10,10
0009    10      CO TINUE
0010            CALL XLOSS
0011            DO 15 J=1,NDAT
0012            AF=CLOSS(J)-DAT(J)
0013            IF(ABS(AF) .LT. .001) GO TO 15
0015            IF(DAT(J) .LT. .0001) GO TO 50
0017            CHISQ=CHISQ+(AF**2)/DAT(J)
0018            GO TO 15
0019    50      CHISQ=CHISQ+10.
0020    15      CONTINUE
0021            NF=NF+1
0022            RETURN
0023    40      CHISQ=1.0E20
0024            RETURN
0025            END
```

```
FORTRAN IV      V01C-03D       THU 16-AUG-79 12:00:33        PAGE 001
CORE=08K, UIC=[100,103]                                ,LP:=OISHLS.ALL

0001          BLOCK DATA
0002          COMMON/ALOSS/CALIB(12,45),DAT(12),CLOSS(12),
             1CNUM1(45),CNUM2(45),CNUM(12),SD(11),TDAT,FLOW,CNCEFF(45)
             2,XDAT,CORFAC(12)
0003          COMMON/STEPIT/X(5),XMAX(5),XMIN(5),DELTAX(5),
             1DELMIN(5),MASK(5),CHISQ,SCHISQ,NF
0004          DATA SD/0.0032,0.0056,0.01,0.0178,0.0316,0.0562,0.1,
             10.178,0.316,0.562,1.0/
0005          DATA DELTAX/.50,.25,.50,.25,.15/
0006          DATA DELMIN/.1,.05,.1,.05,.05/
0007          DATA CNCEFF/45*1.0/
0008          END
```

APPENDIX H

EXPERIMENTAL DATA FOR EAA AND DIFFUSION BATTERY-CNC

Table H1 Aerosol Solution: 0.01% NaCl in water

EAA*		Diffusion Battery-CNC				
			DB: CMS Flow: 6 lpm	DB: CMS Flow: 4 lpm	DB: TSI Flow: 6 lpm	DB: TSI Flow: 4 lpm
Channel No.	I (pA)	Port No.	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)
1	1.000	0	1047	1023	990	1023
2	1.009	1	955	943	909	927
3	1.000	2	887	847	768	724
4	1.036	3	793	740	590	516
5	1.172	4	663	619	415	360
6	0.809	5	559	516	279	233
7	0.274	6	459	418	191	153
8	0.065	7	371	319	127	92.7
9	0.021	8	260	216	79.3	59.5
10	0.007	9	179	146	53.7	35.1
11	0.006	10	109	80.3	32.4	21.4
		11	60.8	43.9		

*Grand Average

**Corrected for coincidence error

Experimental data for EAA and diffusion battery-CNC
(continued)

Table 42 Aerosol Solution: 0.01% DOP in isopropyl alcohol

EAA*		Diffusion Battery-CNC				
			DB: CNS Flow: 6 lpm	DB: CNS Flow: 4 lpm	DB: TSI Flow: 6 lpm	DB: TSI Flow: 4 lpm
Channel No.	I (pA)	Port No.	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)
1	0.047	0	1056	937	868	937
2	0.047	1	981	881	822	896
3	0.047	2	899	822	729	796
4	0.040	3	818	743	583	587
5	0.073	4	734	637	484	429
6	0.463	5	665	540	339	301
7	0.125	6	550	454	235	199
8	0.055	7	444	361	162	129
9	0.020	8	340	227	101	79.3
10	0.004	9	242	137	63	43.6
11	0.003	10	128	85.8	30.4	20.4
		11	79	41.0		

*Grand Average

**Corrected for coincidence error

Experimental Data for EAA and Diffusion Battery-CNC
(continued)

Table XX Aerosol Solution: 0.02% DOP in isopropyl alcohol

EAA			Diffusion Battery-CNC			
			DB: [illegible] Flow: 4 lpm	DB: [illegible] Flow: 4 lpm	DB: [illegible] Flow: 4 lpm	DB: [illegible] Flow: 4 lpm
Channel No.	I (pA)	Run No.	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)
1	1.900	0	928	995	830	132
2	1.460	1	953	946	563	852
3	1.300	2	406	375	848	303
4	1.270	3	618	344	765	593
5	1.385	4	798	710	630	561
6	1.190	5	752	684	719	434
7	0.529*	6	861	668	661	363
8	0.127*	7	948	729	754	296
9	0.041	8	513	417	248	109
10	0.027*	9	413	363	180	123
11	0.017	10	184	279	179	62.4
		11	231	139		

*Grand Average

**Corrected for [illegible] error

Experimental data for EAA and Diffusion Battery-CNC
(continued)

Table A4 Aerosol Solution: 0.1% DOP in isoamyl alcohol

EAA*			Diffusion Battery-CNC			
			DB: 245 Flow: 8 lpm	DB: 245 Flow: 4 lpm	DB: 731 Flow: 8 lpm	DB: 731 Flow: 4 lpm
Channel No.	D (µm)	Port No.	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)	Penetration** (part/cc)
1	[illegible]	0	987	1070	906	896
2	[illegible]	1	796	1001	879	814
3	[illegible]	2	939	963	830	746
4	[illegible]	3	895	954	788	680
5	1.072	4	877	903	685	579
6	1.033	5	839	848	605	504
7	1.280	6	781	780	511	422
8	0.418	7	736	707	438	363
9	0.137	8	640	621	264	271
10	0.084	9	569	514	287	209
11	[illegible]	10	444	388	211	153
		11	329	267		

*Grand Average

**Corrected for coincidence error